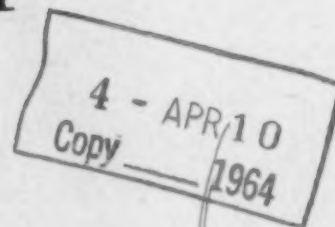




# Radiological Health Data



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VOLUME V, NUMBER 3

MARCH 1964

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service

In August 1959, the President directed the Secretary of Health, Education, and Welfare to intensify Departmental activities in the field of radiological health. The Department was assigned responsibility within the Executive Branch for the collation, analysis and interpretation of data on environmental radiation levels. The Department delegated this responsibility to the Division of Radiological Health, Public Health Service.

*Radiological Health Data* is published by the Public Health Service on a monthly basis. Data are provided to the Division of Radiological Health by other Federal agencies, State health departments, and foreign governments. Pertinent original data and interpretive papers are invited from investigators. Accepted material will be appropriately credited. The reports are reviewed by a Board of Editorial Advisors with representatives from the following Federal agencies:

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# RADIOLOGICAL HEALTH DATA

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# Section I.—Air and Fallout

## FISSION PRODUCT BETA ACTIVITY IN AIRBORNE PARTICULATES AND PRECIPITATION

Early indications of possible increases of fission product activity in various phases of the environment are detectable by continuous surveillance of gross beta activity in air and precipitation. This form of surveillance does not provide sufficient information for assessing human exposure resulting from fallout, but it does form a basis for an alerting system and is useful in determining when and where to conduct more extensive monitoring of radioactivity in food, milk, and water.

Gross beta concentrations in air are presented in reports from the Public Health Service, the Canadian Department of National Health and Welfare, and the Pan American Health Organization.

November 1963 data from three networks were combined and are presented graphically by means of isogram lines on a map of North America (figure 5).

### 1. Radiation Surveillance Network, November 1963

*Division of Radiological Health,  
Public Health Service*

The Radiation Surveillance Network (RSN) is made up of 73 sampling stations distributed throughout the United States (see figure 1). Most of these stations are operated by State health department personnel.

#### *Air*

Airborne particulates are collected continuously on a carbon-loaded cellulose dust filter 4 inches in diameter. A volume of about 1800 cubic meters of air is drawn through the filter during the 24-hour sampling period by a high volume centrifugal blower. Field estimates of the gross beta activity of airborne particulates

are derived by comparing portable survey meter readings of these filters with readings taken from a  $\text{Sr}^{90}\text{-Y}^{90}$  standard. This determination is usually made about 5 hours after the end of the sampling period to eliminate interference from naturally-occurring radon daughters. Until January 24, 1964, the Network's station operators reported their field estimates daily to the Radiation Surveillance Center, Division of Radiological Health, Washington, D. C. Effective on that date, regular reporting of field estimates by telephone was discontinued but provision was made for continued telephone reporting of any values greater than 10 pc/m<sup>3</sup> (or 5 pc/m<sup>3</sup> in Alaska, Hawaii or Puerto Rico).

The filters are then forwarded to the Radiation Surveillance Network laboratory in Rockville, Maryland, for a more refined measurement using a thin-window, gas-flow proportional counter, calibrated with a 38,700 pc  $\text{Sr}^{90}\text{-Y}^{90}$  standard.<sup>1</sup> Each filter is counted at least 3 days after the end of the sampling period and is re-counted 7 days later. The initial 3-day aging of the sample eliminates interference from naturally-occurring radon and thoron daughters. From the two counts, which are separated by the 7-day interval, it is possible to estimate the age of fission products and to extrapolate the activity to the time of collection. The extrapolation is performed by using the Way-Wigner formula:  $AT^{1.2} = C(1)$ .<sup>2</sup> The daily concentrations and estimated age are reported by the PHS in a monthly RSN report (2).

<sup>1</sup> The  $\text{Sr}^{90}\text{-Y}^{90}$  source currently used as a standard was used from April 1962 through August 1963 as 40,000 pc total activity. Beginning with September 1963 data, the nominal activity of the standard was adjusted for decay (about 2½ percent per year) to 38,700 pc.

<sup>2</sup> In this expression, A is the activity, T is the time (in any time unit) after fission product formation, and C is a constant equal to the activity at T=1.



FIGURE 1.—RADIATION SURVEILLANCE NETWORK SAMPLING STATIONS, NOVEMBER 1963

The November 1963 fission-product beta concentrations in surface air (extrapolated to the time of collection) are given in table 1. RSN data are presented with Canadian air data in the form of isogram lines in figure 5.

### Precipitation

Continuous sampling for total precipitation is conducted at most stations on a daily basis, using funnels with collection areas of 0.4 square meter. A 500-ml aliquot of the collected precipitation is evaporated to dryness, and the residue is forwarded to the laboratory for analysis. If the collected sample is between 200 and 500 ml, the entire sample is evaporated. When a sample is smaller than 200 ml (equivalent to 0.5 mm or 0.02 inches of rainfall), the volume of precipitation is reported, but no analysis is made.

In the laboratory the gross beta concentration of precipitation is determined by counting the evaporated sample by the same method used for analyzing the air filters, including the extrapolation to time of collection. Deposition for the sample is determined by:

$$D = \frac{CP}{1000}$$

where  $D$  is the deposition in  $\text{nc}/\text{m}^2$ ,  $C$  is the concentration in  $\text{pc}/\text{liter}$ , and  $P$  is the depth of precipitation in mm. The individual values of deposition and depth of precipitation are totaled for the month, and the average concentration for the month,  $\bar{C}$ , is determined by:

$$\bar{C} = \frac{\sum D}{\sum P} \times 1000$$

The November 1963 average concentrations and total depositions are given in table 2.

### Profiles

The profiles of the monthly average fission product beta activity of airborne particulates for each RSN station covering the period of time from the formation of the network in 1956 to the end of 1960 were published in the July 1961 issue of *Radiological Health Data*. In recent issues, several profiles have been updated each month. The last column of table 1 gives the issue of *Radiological Health Data* having the most recent profile for each station. Eight profiles are updated in figure 2.

TABLE 1.—FISSION PRODUCT GROSS BETA ACTIVITY IN SURFACE AIR, NOVEMBER 1963

[Concentrations in pc/m<sup>3</sup>]

Station location	Number of samples	Maximum	Minimum	Average <sup>a</sup>	Last profile in RHD
Alaska: Adak	30	1.5	<0.10	0.50	Nov. 1963
Anchorage	28	2.1	0.20	0.95	Jul. 1963
Attu	29	1.7	<0.10	0.72	Dec. 1963
Fairbanks	16	0.57	0.22	0.32	Aug. 1963
Juneau	23	1.0	<0.10	0.39	Sep. 1963
Kodiak	21	1.7	0.25	0.71	Oct. 1963
Nome	20	1.7	0.16	0.69	Feb. 1964
Point Barrow	23	0.71	<0.10	0.39	Jan. 1964
St. Paul Island	26	1.7	0.15	0.78	Mar. 1964
Ariz: Phoenix	24	3.5	0.64	1.4	Sep. 1963
Ark: Little Rock	27	2.0	0.68	1.4	Sep. 1963
Calif: Berkeley	19	1.4	0.38	0.75	Oct. 1963
Los Angeles	17	2.9	0.44	1.5	Feb. 1964
Canal Z: Ancon	12	0.62	<0.10	<0.19	b
Colo: Denver	27	2.7	0.49	1.3	Nov. 1963
Conn: Hartford	25	1.6	<0.10	0.82	Oct. 1963
Del: Dover	13	2.2	0.44	1.1	Aug. 1963
D. C: Washington	30	3.1	<0.10	1.1	Mar. 1963
Fla: Jacksonville	26	2.2	0.21	1.3	Oct. 1963
Miami	30	2.8	0.16	1.4	Feb. 1964
Ga: Atlanta	25	6.6	0.77	2.0	Jul. 1963
Guam: Agana	30	0.9	<0.10	0.37	Mar. 1963
Hawaii: Honolulu	25	1.5	0.21	0.72	Feb. 1964
Idaho: Boise	27	2.4	<0.10	0.88	Dec. 1963
Ill: Springfield	26	1.3	0.14	0.81	Mar. 1964
Ind: Indianapolis	23	1.9	0.15	1.0	Jul. 1963
Iowa: Iowa City	28	1.6	0.32	0.88	Jan. 1964
Kans: Topeka	24	1.9	0.53	1.2	Jul. 1963
Ky: Frankfort	26	2.2	0.23	0.98	Feb. 1964
La: New Orleans	24	2.6	0.53	1.2	Mar. 1964
Maine: Augusta	30	1.7	<0.10	0.71	Mar. 1964
Presque Isle	28	1.0	<0.10	0.43	Nov. 1963
Md: Baltimore	19	2.6	<0.10	1.1	Nov. 1963
Rockville	15	3.2	<0.10	1.4	Mar. 1964
Mass: Lawrence	29	1.7	<0.10	0.83	Aug. 1963
Winchester	24	1.3	<0.10	0.65	b
Mich: Lansing	30	2.2	0.15	1.1	Feb. 1964
Minn: Minneapolis	22	1.4	0.21	0.80	Mar. 1964
Miss: Jackson	28	2.5	0.53	1.3	Mar. 1963
Pascagoula	8	2.1	0.59	1.6	Dec. 1963
Mo: Jefferson City	25	1.6	0.33	1.0	Nov. 1963
Mont: Helena	23	2.3	0.61	1.2	Nov. 1963
Neb: Lincoln	1	0.57	0.57	0.57	Apr. 1963
Nev: Las Vegas	23	5.1	0.82	2.4	Jul. 1963
N.H: Concord	17	1.8	0.11	1.0	Feb. 1964
N.J: Trenton	29	2.0	<0.10	0.98	Apr. 1964
N.Mex: Santa Fe	21	5.6	0.66	1.6	Dec. 1963
N.Y: Albany	17	2.0	<0.10	0.94	Jul. 1963
Buffalo	18	2.5	<0.10	0.85	Nov. 1963
New York	12	1.6	0.14	0.84	Dec. 1963
N.C: Gastonia	25	1.8	<0.10	1.1	Jan. 1964
N.Dak: Bismarck	24	2.0	0.34	0.91	Feb. 1964
Ohio: Cincinnati	18	1.5	<0.10	0.90	Aug. 1963
Columbus	27	2.1	<0.10	1.0	Feb. 1963
Painesville	28	3.2	0.20	1.3	Oct. 1963
Okla: Oklahoma City	24	1.4	0.28	1.0	Apr. 1963
Ponca City	23	1.1	0.33	0.69	Oct. 1963
Ore: Portland	27	3.1	0.40	1.3	Oct. 1963
Pa: Harrisburg	21	2.6	<0.10	0.86	Jan. 1964
P.R: San Juan	23	0.8	0.14	0.39	Mar. 1964
R.I: Providence	25	1.9	<0.10	0.96	Jan. 1964
S.C: Columbia	24	1.5	0.16	0.97	Dec. 1963
S.Dak: Pierre	29	1.8	0.36	0.97	Sep. 1963
Tenn: Nashville	30	3.2	0.25	1.3	Jan. 1964
Tex: Austin	25	2.3	0.75	1.4	Aug. 1963
El Paso	29	2.5	0.55	1.3	Jan. 1964
Utah: Salt Lake City	30	2.6	0.41	1.2	Aug. 1963
Vt: Barre	27	2.2	<0.10	0.87	Sep. 1963
Va: Richmond	27	1.7	<0.10	0.84	Sep. 1963
Wash: Seattle	29	1.0	<0.10	0.48	Jul. 1963
W.Va: Charleston	28	2.0	0.13	0.96	Dec. 1963
Wisc: Madison	26	1.4	0.14	0.85	Sep. 1963
Wyo: Cheyenne	26	2.8	0.52	1.2	Aug. 1963
Network summary	1738	6.6	<0.10	0.98	

<sup>a</sup> The monthly average is calculated by weighting the individual samples with length of sampling period. Values of <0.10 are assumed to be 0.10 for averaging purposes. If the <0.10 values represent more than 10% of the average, a less-than sign is placed in front of the average.

<sup>b</sup> Initial profile scheduled for a future issue.

TABLE 2.—FISSION PRODUCT GROSS BETA ACTIVITY IN PRECIPITATION, NOVEMBER 1963

Station location	Average concentration <sup>a</sup> (pc/liter)	Total deposition <sup>a</sup> (nc/m <sup>2</sup> )
Alaska: Anchorage	280	0.5
Fairbanks	1,200	17
Juneau	450	71
Ark: Little Rock	350	52
Calif: Berkeley	230	18
Los Angeles	200	14
Colo: Denver	<200	<3
Conn: Hartford	220	19
D.C: Washington	310	41
Fla: Jacksonville	<200	<13
Miami	350	22
Ga: Atlanta	b	b
Idaho: Boise	b	b
Ill: Springfield	380	8
Ind: Indianapolis	420	28
Iowa: Iowa City	460	20
Kans: Topeka	630	15
Ky: Frankfort	670	20
La: New Orleans	<200	<53
Maine: Augusta	320	93
Presque Isle	540	11
Md: Baltimore	460	183
Mass: Lawrence	450	100
Winchester	480	47
Mich: Lansing	440	24
Minn: Minneapolis	350	5
Miss: Jackson	<200	<117
Mo: Jefferson City	320	13
Mont: Helena	8,000	18
Neb: Lincoln	b	b
Nev: Las Vegas	b	b
N.J: Trenton	280	8
N.Mex: Santa Fe	500	7
N.Y: Albany	240	21
Buffalo	b	b
N.C: Gastonia	<200	<19
N.Dak: Bismarck	b	b
Ohio: Columbus	1,000	13
Painesville	370	41
Okla: Oklahoma City	b	b
Ponca City	300	15
Ore: Portland	270	32
Pa: Harrisburg	280	22
P.R: San Juan	<200	<18
R.I: Providence	380	27
S.C: Columbia	280	39
S.Dak: Pierre	b	b
Tenn: Nashville	550	29
Tex: Austin	680	2
El Paso	1,000	14
Utah: Salt Lake City	230	9
Vt: Barre	490	41
Va: Richmond	220	29
Wash: Seattle	300	48
W.Va: Charleston	370	27
Wisc: Madison	340	18
Wyo: Cheyenne	b	b

<sup>a</sup> The minimum concentration reported for a single sample is 200 pc/liter. If the individual sample has a concentration of <200 pc/liter, the deposition for that sample is calculated by  $D = <0.2 P$  in nc/m<sup>2</sup> (see text). A less-than sign (<) is used with the monthly total deposition if the sum of the individual less-than values represents more than 10 percent of the total. The monthly average concentration is then calculated as described in text, retaining the less-than sign when used with the total deposition.

<sup>b</sup> No evaporated sample received.

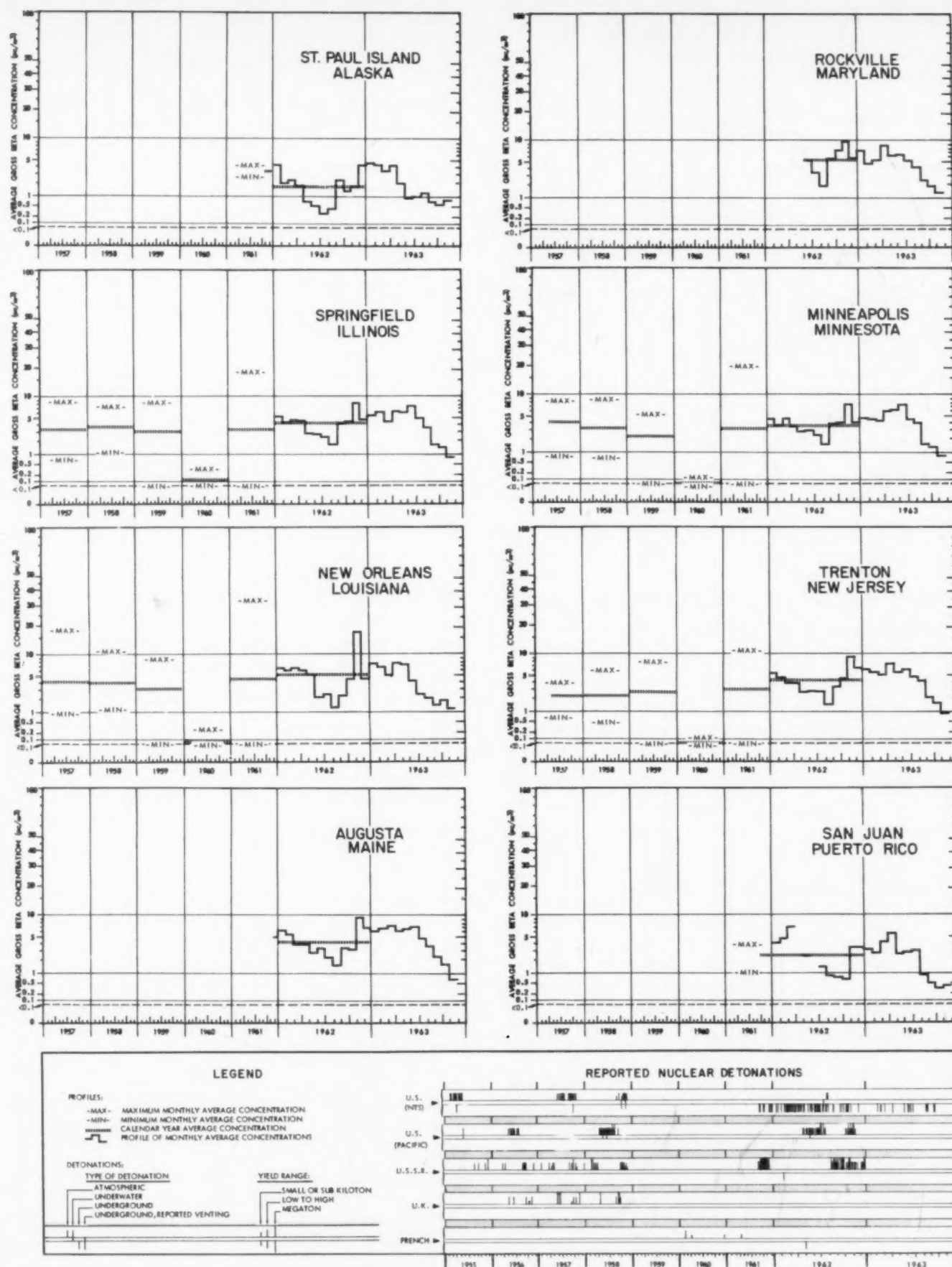


FIGURE 2.—MONTHLY AND YEARLY PROFILES OF BETA ACTIVITY IN AIR, RADIATION SURVEILLANCE NETWORK, 1957—NOVEMBER 1963

## 2. Canadian Air Monitoring Program,<sup>3</sup> November 1963

Department of National Health and Welfare,  
Ottawa, Canada

As part of its Radioactive Fallout Study Program, the Radiation Protection Division of the Canadian Department of National Health and Welfare monitors air and precipitation. Twenty-four collection stations are located at airports (see figure 3), where the sampling equipment is operated by personnel from the Meteorological Services Branch of the Department of Transport. Detailed discussions of the sampling procedures, methods of analysis, and interpretation of results of the radioactive fallout program are contained in reports of the Department of National Health and Welfare (3-7).

### Air

Each air sample involves the collection of particulates from about 650 cubic meters of air drawn through a high-efficiency 4-inch-diameter filter during a 24-hour period. These filters

are sent daily to the Radiation Protection Division Laboratory in Ottawa. At the laboratory, a 2-inch-diameter disk is cut from each filter and counted with a thin-end-window, gas flow Geiger-Mueller counter system, calibrated with a  $\text{Sr}^{90}\text{-Y}^{90}$  standard. Four successive measurements are made on each filter to permit correction for natural activities and for the decay of short-lived fission products. The results are extrapolated to the end of the sampling period. Canadian air data for November 1963 are given in table 3 and presented in conjunction with U. S. and Mexico data by an isogram map (figure 5).

### Precipitation

The amount of radioactive fallout deposited on the ground is determined from measurements on material collected in special polyethylene-lined rainfall pots. The collection period for each sample is one month. After transfer of the water to the sample container, the polyethylene liner is removed, packed with the sample, and sent to the laboratory.

Strontium and cesium carriers are added to all samples on arrival at the laboratory. Other carriers are also added to selected samples according to the specific radio-nuclides to be determined. The samples are then filtered and the filtrate evaporated to near dryness. The

<sup>3</sup> Data from *Radiation Protection Programs, Vol. 1, No. 12*: 11-24, Radiation Protection Division, Canadian Department of National Health and Welfare, (December 1963).



FIGURE 3.—CANADIAN AIR AND PRECIPITATION SAMPLING STATIONS, NOVEMBER 1963

TABLE 3.—FISSION PRODUCT GROSS BETA ACTIVITY IN AIR, CANADA, NOVEMBER 1963

[Average concentrations in pc/m<sup>3</sup>]

Station	Number of samples	Maximum	Minimum	Average
Calgary.....	27	3.0	0.3	1.0
Coral Harbour.....	23	2.0	0.1	0.5
Edmonton.....	29	1.4	0.4	0.7
Ft. Churchill.....	30	1.6	0.1	0.6
Ft. William.....	29	1.7	0.0	0.9
Fredericton.....	29	1.4	0.0	0.5
Goose Bay.....	27	1.3	0.0	0.6
Halifax.....	28	2.1	0.1	0.9
Inuvik.....	30	1.4	0.3	0.7
Montreal.....	30	1.8	0.1	0.7
Moosonee.....	30	2.1	0.0	0.6
Ottawa.....	24	2.0	0.1	0.8
Quebec.....	29	1.7	0.1	0.6
Regina.....	30	4.8	0.3	1.0
Resolute.....	30	1.0	0.0	0.5
St. John's, Nfld.....	29	2.3	0.1	0.8
Saskatoon.....	30	3.5	0.4	1.0
Sault Ste. Marie.....	29	1.9	0.0	0.9
Toronto.....	30	1.0	0.0	0.4
Vancouver.....	30	1.8	0.1	0.7
Whitehorse.....	30	1.8	0.2	0.7
Windsor.....	30	2.6	0.1	1.2
Winnipeg.....	30	1.9	0.3	0.9
Yellowknife.....	29	1.1	0.1	0.6
Network summary.....	692	4.8	0.0	0.7

filter paper containing insoluble matter is ignited together with the polyethylene liner at 450°C. The ash is combined with the soluble fraction, transferred to a glass planchet, evaporated under an infra-red lamp and then counted with a thin-end-window Geiger-Mueller counter

TABLE 4.—FISSION PRODUCT GROSS BETA ACTIVITY IN PRECIPITATION, CANADA, NOVEMBER 1963

Station	Total beta activity	
	pc/liter	nc/m <sup>2</sup>
Calgary.....	761	10.0
Coral Harbour.....	547	8.2
Edmonton.....	298	6.7
Ft. Churchill.....	426	22.7
Ft. William.....	758	27.7
Fredericton.....	246	44.4
Goose Bay.....	268	20.9
Halifax.....	371	58.5
Inuvik.....	276	5.3
Montreal.....	394	58.7
Moosonee.....	329	27.4
Ottawa.....	259	42.9
Quebec.....	395	55.9
Regina.....	1453	8.1
Resolute.....	234	18.4
St. John's, Nfld.....	271	36.4
Saskatoon.....	325	10.2
Sault Ste. Marie.....	385	39.4
Toronto.....	409	24.6
Vancouver.....	322	52.5
Whitehorse.....	211	5.4
Windsor.....	809	29.0
Winnipeg.....	390	12.5
Yellowknife.....	197	7.0
Average.....	431	26.4

calibrated with a Sr<sup>90</sup>-Y<sup>90</sup> source. Gross beta activities for November samples are given in table 4. Radionuclide analyses appear quarterly.

### 3. Mexican Air Monitoring Program, November 1963

#### National Commission of Nuclear Energy Mexico

The Radiation Surveillance Network of Mexico was established by the Comision Nacional de Energia Nuclear (CNEN) through its Radiological Protection Program (RPP) in 1961. The network is made up of 17 stations (see figure 4), twelve of which are located at airports and operated by airline personnel. The remaining five stations are located at Mexico City, Mérida, Veracruz, San Luis Potosí and Ensenada. Staff members of the RPP operate the station at Mexico City while the other four stations are manned by members of the Centro de Previsión del Golfo de México, the Chemistry Department of the University of Mérida, the Instituto de Zonas Deserticas of the University of San Luis Potosí, and the Escuela Superior de Ciencias Marinas of the University of Baja California, respectively.



FIGURE 4.—FALLOUT NETWORK SAMPLING STATIONS IN MEXICO

#### Sampling

The sampling procedure involves drawing air for 24 hours a day, 3 or more days a week, at the rate of approximately 1,200 cubic meters

TABLE 5.—GROSS BETA ACTIVITY OF AIRBORNE PARTICULATES, MEXICO, NOVEMBER 1963

[Concentrations in pc/m <sup>3</sup> ]				
Station	Number of samples	Maximum	Minimum	Average
Acapulco.....	8	1.3	0.3	0.8
Ciudad Juárez.....	23	2.2	0.2	1.1
Chihuahua.....	23	2.9	0.5	1.2
Ensenada.....	8	2.3	0.5	0.9
Guadalajara <sup>a</sup> .....	3	0.5	0.1	—
Guaymas.....	3	3.5	0.9	—
La Paz.....	14	2.6	0.9	1.4
Matamoros.....	7	1.0	0.5	0.8
Mazatlán <sup>a</sup> .....	1	1.5	—	—
Mérida.....	14	2.1	0.4	1.0
México, D.F. <sup>b</sup> .....	18	2.1	0.1	0.6
Nuevo Laredo.....	4	1.3	0.7	—
San Luis Potosí.....	7	1.3	0.1	0.6
Tampico.....	12	1.9	0.6	1.1
Torreón.....	15	2.7	0.7	1.3
Tuxtla Gutiérrez <sup>a</sup> .....	0	—	—	—
Veracruz.....	3	0.6	0.3	—

<sup>a</sup> Sampling equipment was out of order for all or most of the month, therefore, the average is not given.

<sup>b</sup> Mexico City.

per day, through a 6 x 8 inch high-efficiency glass fiber filter using high volume samplers. After each 24-hour sampling period, the filter is removed and forwarded via air mail to the "Laboratorio de Estudios Sobre Contaminación Radiactiva", CNEN, in Mexico City for assay of gross beta activity. A minimum of 3 days after collection is allowed for decay of radon and thoron daughter natural radioactivity. Data are not extrapolated to time of collection.

The maximum, minimum and average fission product beta concentrations in surface air during November 1963 are presented in table 5. The data are also represented after adjustment for intercalibration in the beta activity isogram map of North America, figure 5.

#### 4. Pan American Air Sampling Program, November 1963

##### *Pan American Health Organization and Public Health Service*

Gross beta activity in air is monitored by three countries in South America under the auspices of a collaborative program developed by the Pan American Health Organization and the Public Health Service (PHS) for assisting countries of the Americas to develop their own radiological health programs. The sampling equipment and analytical services are provided by the Division of Radiological Health, PHS,

TABLE 6.—GROSS BETA ACTIVITY IN AIR, NOVEMBER 1963

[Concentrations in pc/m <sup>3</sup> ]				
Sampling stations	Number of samples	Maximum	Minimum	Average <sup>a</sup>
Caracas, Venezuela.....	21	0.50	<0.10	0.18
Lima, Peru.....	14	0.23	<0.10	0.13
Santiago, Chile.....	16	0.20	<0.10	0.13

<sup>a</sup> The monthly average is calculated by weighting the individual samples with length of sampling period. Values of <0.10 are assumed to be 0.10 for averaging purposes. If the <0.10 values represent more than 10 percent of the average, a less-than sign is placed in front of the average.

and are identical with those employed for the Radiation Surveillance Network.

The three air sampling stations included in the program are operated by the technical staff of the ministry of health in each country. The station in Santiago, Chile is operated by the Occupational Health Service; in Lima, Peru by the Institute of Occupational Health; and in Caracas, Venezuela by the Venezuelan Institute for Scientific Investigations. The Caracas station began operation in November 1962 and the other two stations were started the following month.

The November 1963 air monitoring results from the three participating countries are given in table 6. The Caracas station is shown on the gross beta concentration isogram map (figure 4). The November average at this station, adjusted by the RSN intercalibration factor is <0.14 pc/m<sup>3</sup>, which is considerably below the lowest isogram lined used on the map (1 pc/m<sup>3</sup>).<sup>4</sup>

#### 5. Gross Beta Activity in Air, North America, November 1963

Beginning with January 1963 data, monthly average concentrations of airborne gross beta activity in Canada and the United States have been presented in combined form as isogram maps of most of North America.<sup>5</sup> The data from the Radiation Surveillance Network and the Canadian Air Network were adjusted to each other by means of an intercalibration factor derived by Lockhart and Patterson (8).

With the formation of the Mexican Air monitoring program, new intercalibration ratios were determined, this time including the Canadian Air Monitoring Program, Radiation Sur-

<sup>4</sup> The RSN factor is 1.28 (see page 112).

<sup>5</sup> The January to October 1963 Isogram Maps were published in the May 1963 through February 1964 *Radiological Health Data*.

veillance Network, National Air Sampling Network, the new 80th Meridian Network, and the Mexican Air Monitoring Program (9). The new intercalibration factors include some changes in standardization in both the RSN and the Canadian Air Network, effective September 1963. The intercalibration factors are, therefore, not the same as were previously used.

Figure 5 shows the November 1963 activity in air throughout North America based on the data from the Canadian Air Monitoring Program, the Radiation Surveillance Network and the Mexican Air Monitoring Program. An intercalibration factor of 1.28 was applied to the RSN data and 0.81 to the Mexican data thereby using Canadian data as a frame of reference.

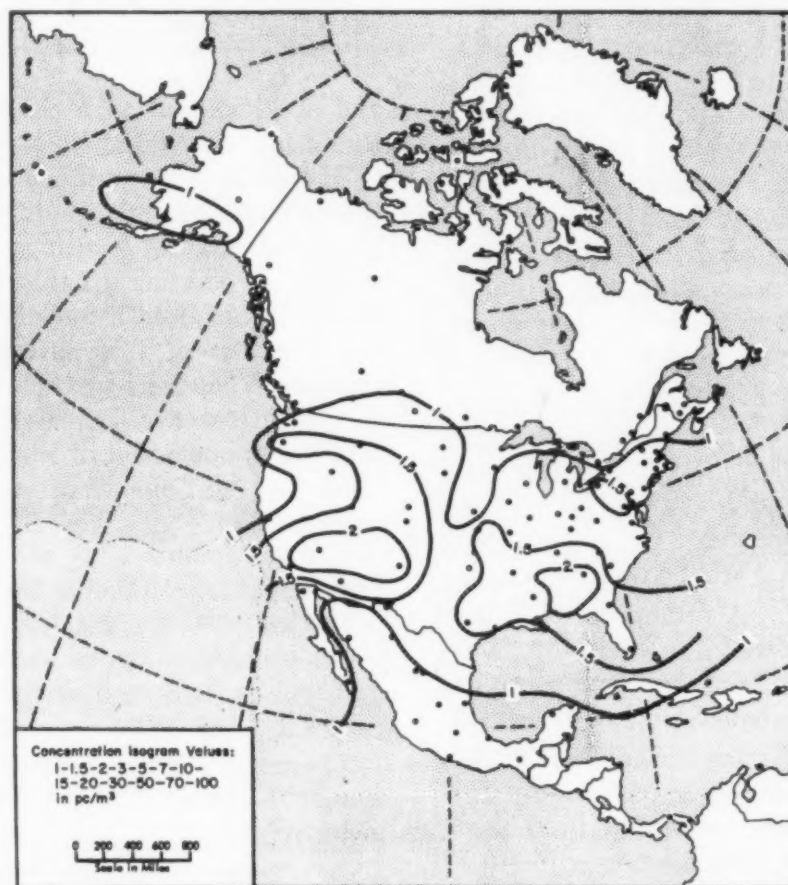


FIGURE 5.—ISOGRAMS OF AVERAGE GROSS BETA CONCENTRATIONS IN AIR THROUGHOUT NORTH AMERICA, NOVEMBER 1963

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# FISSION PRODUCT GAMMA ACTIVITY IN AIR— 80TH MERIDIAN NETWORK, OCTOBER 1963

Health and Safety Laboratory, Atomic Energy  
Commission

Total gamma activity measurements for weekly ground-level air filter samples taken at 80th Meridian stations (see figure 1) during October 1963 are listed in table 1, together with monthly average concentrations and the fractions of the total gamma activity in excess of 1 Mev. The monthly average total gamma concentrations are plotted in figure 2, as an activity-latitude profile and compared with the third quarter average profile. Details of sampling procedures and analytical methodology have previously been given by Collins (1, 2).

The October results in general show no major changes in ground level air activity concentrations in either the Northern or the Southern Hemisphere to results obtained for

September 1963. The October average for the northern sites was 0.93 photons per minute per cubic meter ( $\gamma/\text{min}/\text{m}^3$ ) with a range of from 0.12 to 1.9  $\gamma/\text{min}/\text{m}^3$ . At stations in the South-

TABLE 1.—ACTIVITY OF SURFACE AIR, 80TH  
MERIDIAN NETWORK, OCTOBER 1963

Sampling station	Sampling period (dates— noon to noon)	Gamma activity (photons/min/m <sup>3</sup> )		Gamma ratio ( $\gamma > 1 \text{ Mev}$ total $\gamma$ )
		Filter	Average for month	
Thule.....	10/1-8	0.453	0.814	0.042
	10/8-15	0.814		0.042
	10/15-22	1.07		0.041
	10/22-11/1	0.917		0.041
Moosonee.....	10/1-8	0.834	0.961	0.048
	10/8-15	1.15		0.040
	10/15-22	0.865		0.038
	10/22-11/1	0.996		0.039
New York.....	10/1-8	0.216	1.28	0.049
	10/8-15	1.33		0.049
	10/15-22	0.339		0.044
Washington.....	10/1-8	1.71	1.55	0.038
	10/8-15	1.42		0.049
	10/15-22	1.73		0.048
	10/22-11/1	1.35		0.047
Miami.....	10/1-8	2.42	1.88	0.049
	10/8-15	1.93		0.048
	10/15-22	1.48		0.049
	10/22-11/1	1.69		0.045
Mauna Loa.....	10/1-8	0.406	0.574	0.046
	10/8-15	0.864		0.048
	10/15-22	0.531		0.045
	10/22-11/1	0.496		0.043
San Juan.....	10/1-8	0.157	0.278	0.051
	10/8-15	0.192		0.052
	10/15-22	0.516		0.038
	10/22-11/1	0.248		0.044
Miraflores.....	10/1-8	0.134	0.122	0.055
	10/8-15	0.0424		0.071
	10/15-22	0.0710		0.067
	10/22-11/1	0.240		0.048
Guayaquil.....	10/1-8	0.123	0.101	0.046
	10/8-15	0.0561		0.072
	10/15-22	0.153		0.045
	10/22-11/1	0.0724		0.051
Lima.....	10/1-8	0.154	0.144	0.054
	10/8-15	0.140		0.043
	10/15-22	0.112		0.052
	10/22-11/1	0.168		0.042
Chacaltaya.....	10/1-8	0.113	0.0874	0.043
	10/8-15	0.0691		0.061
	10/15-22	0.0818		0.026
	10/22-11/1	0.0855		0.051
Antofagasta.....	10/1-8	0.139	0.109	0.051
	10/8-15	0.0836		0.054
	10/15-22	0.120		0.041
	10/22-11/1	0.0917		0.051
Santiago.....	10/1-8	0.0962	0.0784	0.055
	10/8-15	0.0602		0.063
	10/15-22	0.0780		0.053
	10/22-11/1	0.0790		0.055
Puerto Montt.....	10/1-8	0.0284	0.0308	0.010
	10/8-15	0.0233		0.023
	10/15-22	0.0251		0.041
	10/22-11/1	0.0465		0.044
Punta Arenas.....	10/8-15	0.0455	0.0278	0.028
	10/15-22	0.0191		0.035
	10/22-11/1	0.0187		0.052

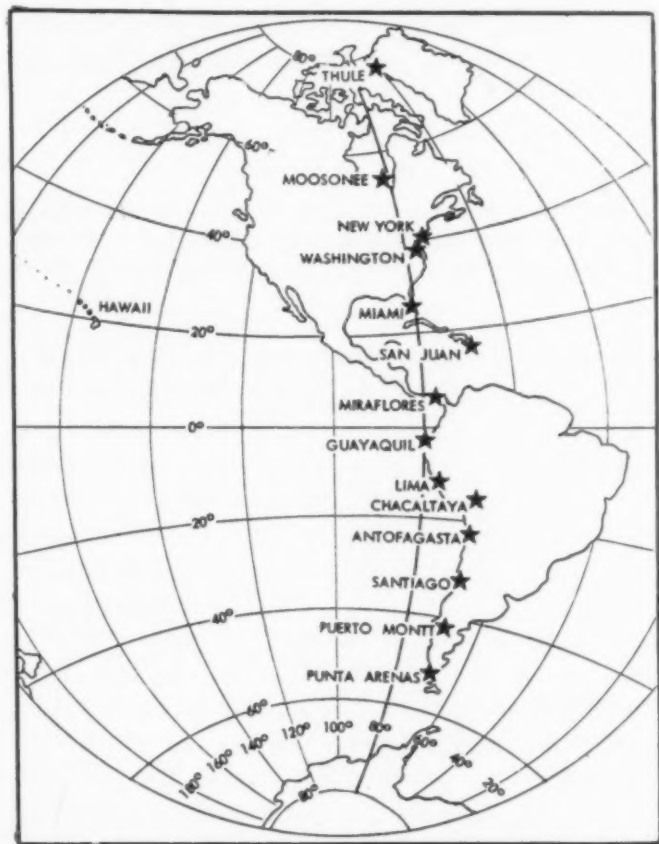


FIGURE 1.—80TH MERIDIAN NETWORK SAMPLING  
STATIONS

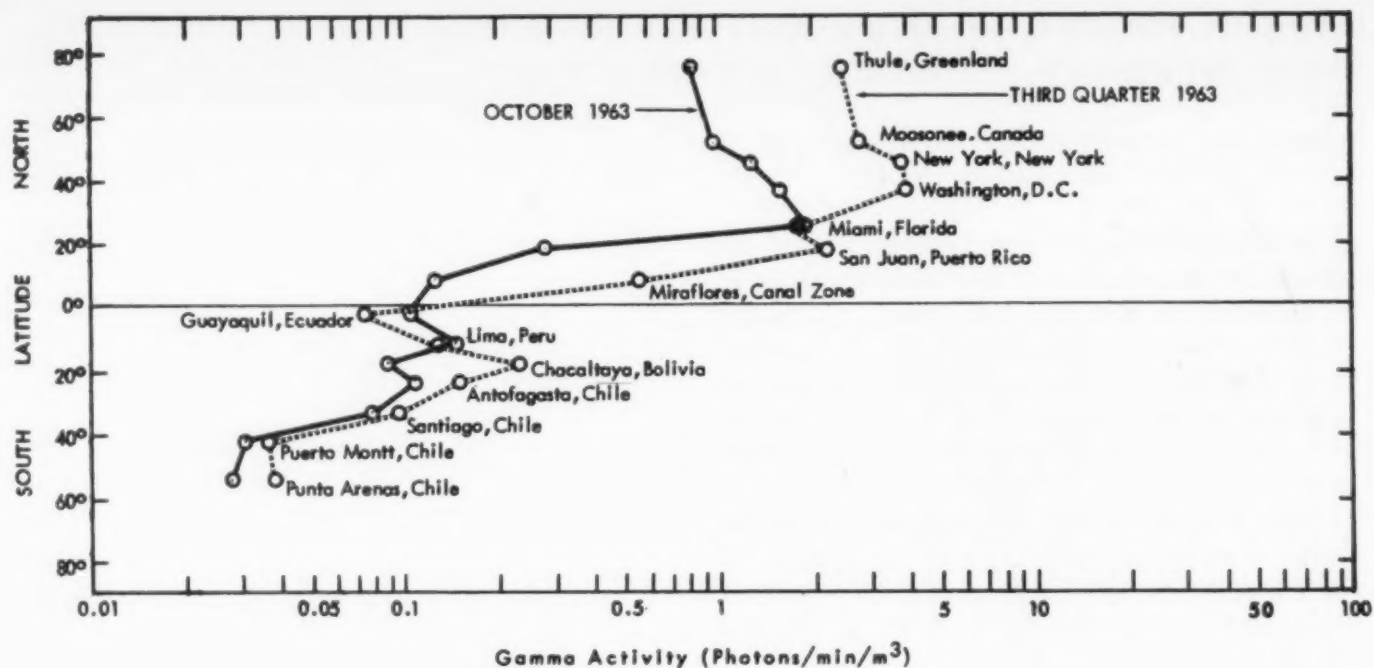


FIGURE 2.—PROFILE OF SURFACE AIR GAMMA ACTIVITY, THIRD QUARTER AND NOVEMBER 1963

ern Hemisphere the average concentration was 0.083 with a range of from 0.028 to 0.14  $\gamma/\text{min}/\text{m}^3$ .

Total beta activity estimates have not been given for samples taken since June 1963 because of the continued appearance of larger concentrations of high energy gamma emitters than would be expected in old fission product mixtures. According to observations reported for thermally-irradiated  $\text{U}^{235}$  (1) only about 1 percent of the total gamma activity in the October samples would be expected to be in excess of 1 Mev. The actual values average 4.7 percent and 4.6 percent for the Northern and Southern Hemispheres respectively. Continued

radiochemical and gamma spectrometric analyses are in process to determine to what extent enrichment in high-energy activation products such as  $\text{Y}^{88}$  and  $\text{Sb}^{124}$  and fission products such as  $\text{Ce}^{144}$  and  $\text{Ru}^{106}$  is responsible for these discrepancies.

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## Section II.—Milk and Food

### MILK SURVEILLANCE

Although milk is only one of the many sources of dietary intake of radionuclides, it is the single food item most often used as an indicator of the population's intake of radionuclides from the environment. This is because fresh milk is consumed by a large segment of the United States population and contains most of the radionuclides occurring in the environment which have been identified as being biologically important. In addition, milk is produced and consumed on a regular basis, is convenient to handle, is easily analyzed, and samples which are representative of milk consumption in any area can be readily obtained.

#### 1. Pasteurized Milk Network, November 1963

*Division of Radiological Health and Division of Environmental Engineering and Food Protection, Public Health Service*

The Public Health Service pasteurized milk radionuclide surveillance program had its origin in a raw milk monitoring network (1) established by the Service in 1957. One of the primary objectives of this network was the development of methods for milk collection and radiochemical analysis suitable for larger scale programs.

Experience derived from this earlier network led to the activation of a pasturized milk sampling program with stations selected to provide nationwide surveillance of milk production and consumption areas. The present network, which consists of 63 stations, has at least one station in every State, the Canal Zone, and Puerto Rico.

Through the cooperation of State and local

milk sanitation authorities, samples are routinely collected at each of these stations. Composites of the samples are preserved with formaldehyde and are sent to the PHS Southwestern (SWRHL), Southeastern (SERHL), or Northeastern Radiological Health Laboratories (NERHL) for analysis. Gamma analyses for iodine-131 are made within 3 to 6 days after sample collection, and any results exceeding 100 pc/liter are immediately telephoned to State health officials for possible public health action. Complete analytical results are available 6 to 7 weeks after collection; publication in *Radiological Health Data* follows 3 to 4 months after sample collection.

#### *Sampling and Compositing Procedures*

The method specifies that each station's sample be composited of subsamples from each milk processing plant in proportion to the plant's average sales in the community served. At most stations the composited sample represents from 80 to 100 percent of the milk processed. Prior to September 15, 1961, the composite sample was taken from one day's sales per month and was as representative of the community's supply as could be achieved under practical conditions. Beginning with the resumption of nuclear weapons testing in the atmosphere in September 1961, and continuing through January 1963, samples were collected twice a week at nearly all stations and daily for short periods at selected stations. Since then, the sampling frequency has been reduced to once a week.

#### *Analytical Errors*

Iodine-131, cesium-137 and barium-140 concentrations are determined by gamma scintil-

lation spectroscopy.<sup>1</sup> After gamma scanning, a two-week composite is analyzed radiochemically for strontium-89 and strontium-90. There is an inherent statistical variation associated with all measurements of radionuclide concentrations. With the low radionuclide levels which are usually found in milk and other environmental samples, this variation on a percentage basis is relatively high. The variation is dependent upon such factors as the method of chemical analysis, the sample counting rate and counting time, interferences from other radionuclides, and the background count. For milk samples, counting times of 50 minutes for gamma spectroscopy and 30 to 50 minutes for low background beta determinations are used. Table 1 shows the approximate total analytical error (including counting error) associated with radionuclide concentrations in milk. The  $\pm 2\sigma$  range about the measured concentration corresponds to a 95 percent certainty that the true concentration is within this range. The minimum detectable concentration is defined as the measured concentration at which the two-standard-deviation analytical error is equal to the measurement. Accordingly, the minimum detectable concentrations in units of pc/liter are Sr<sup>89</sup>, 5; Sr<sup>90</sup>, 2; Cs<sup>137</sup>, 10; Ba<sup>140</sup>, 10; and I<sup>131</sup>, 10. At these levels and below, the counting error comprises nearly all of the analytical error.

<sup>1</sup> Southeastern Radiological Health Laboratory employs a radiochemical procedure for barium-140 analysis.

TABLE 1.—ANALYTICAL ERRORS ASSOCIATED WITH ESTIMATED CONCENTRATIONS FOR SELECTED RADIONUCLIDES IN MILK

Nuclide	Estimated concentration (pc/liter)	Error <sup>a</sup> (pc/liter)	Estimated concentration (pc/liter)	Error <sup>a</sup> (percent of concentration)
Iodine-131	0 to 100	$\pm 10$	100 or greater	$\pm 10\%$
Barium-140	0 to 100	$\pm 10$	100 or greater	$\pm 10\%$
Cesium-137	0 to 100	$\pm 10$	100 or greater	$\pm 10\%$
Strontium-89	0 to 50	$\pm 5$	50 or greater	$\pm 10\%$
Strontium-90	0 to 20	$\pm 2$	20 or greater	$\pm 10\%$

<sup>a</sup> Two standard deviations.

Calcium analyses at SERHL are done by an ion exchange and volumetric method while at NERHL and SWRHL an ethylenediaminetetraacetic acid (EDTA) method is used. Stable potassium concentrations are estimated from the potassium-40 concentrations determined from the gamma spectrum.

#### Data Presentation

Table 2 presents summaries of the analyses for November 1963 (October 27–November 30, 1963). Although not shown in table 2, the iodine-131 and barium-140 monthly average concentrations in milk were  $<10$  pc/liter. When a radionuclide is reported by a laboratory as being below the minimum detectable concentration, one-half the minimum detectable value is used as the best approximation in calculating the monthly average. Beginning with October 1963 data, however, zero has been used as the best approximation to a nondetectable concentration of iodine-131 or barium-140. A similar procedure is used for the network average.

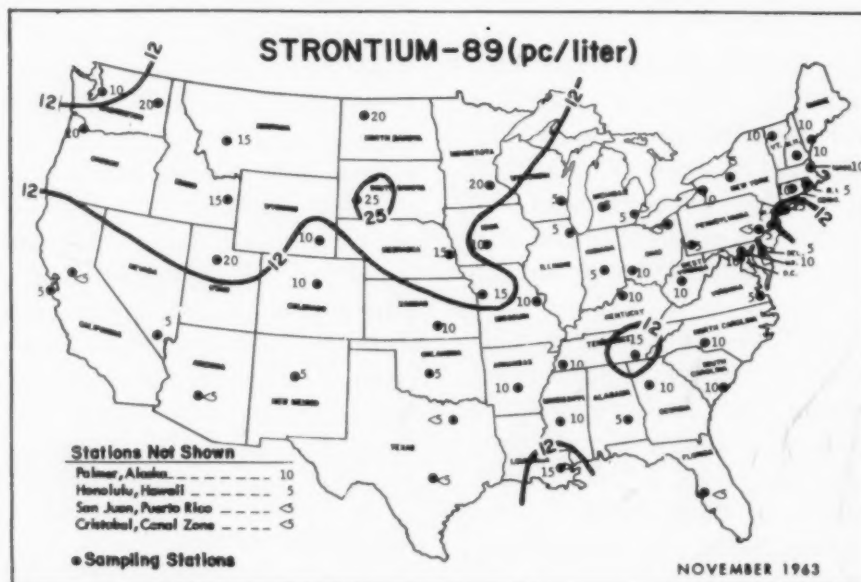


FIGURE 1.—STRONTIUM-89 CONCENTRATIONS IN PASTEURIZED MILK

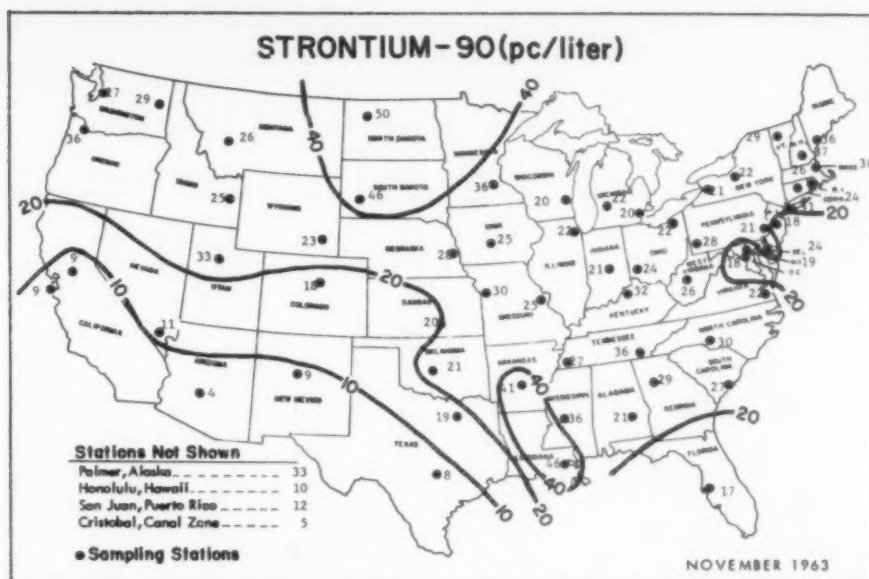


FIGURE 2.—STRONTIUM-90 CONCENTRATIONS IN PASTEURIZED MILK

Figures 1, 2, and 3 are isoconcentration maps showing the estimated strontium-89, strontium-90, and cesium-137 concentrations in milk over the entire country. The value printed beside each station is the monthly average concentration for that station. The isoconcentration maps were developed by arbitrary interpolation between values for the individual stations. Additional modifications to the isoconcentration contours are made according to available information on milksheds. In order to develop the distribution of the network's stations versus radionuclide concentra-

tions in milk, table 3 has been prepared using monthly average data shown in table 2.

Continuing the practice followed in previous issues of *RHD* the average monthly strontium-90 concentrations in pasteurized milk from 16 selected cities in the sampling program are presented in figure 4. Each graph shows the strontium-90 concentrations in milk from one city in each of the four U. S. Bureau of Census regions. This method of selection permits graphic presentation of data for each city in the network three times a year.

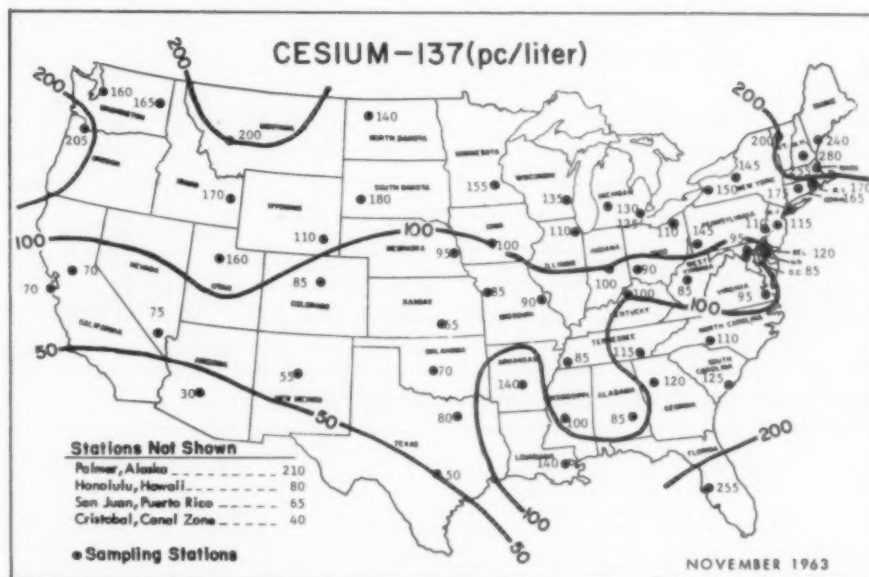


FIGURE 3.—CESIUM-137 CONCENTRATIONS IN PASTEURIZED MILK

TABLE 2.—RADIOACTIVITY IN PASTEURIZED MILK, NOVEMBER 1963\*

[Average radioactivity concentrations in pc/liter]

Sampling locations		Calcium (g/liter)		Potassium (g/liter)		Strontium-89		Strontium-90		Cesium-137		Last Sr <sup>90</sup> graph in RHD
		Third quarter	Avg. for month	Third quarter	Avg. for month	Third quarter	Avg. for month	Third quarter	Avg. for month	Third quarter	Avg. for month	
Ala:	Montgomery	1.17	1.22	1.4	1.5	45	5	21	21	85	85	Feb. 64
Alaska:	Palmer	1.17	1.25	1.5	1.6	45	10	24	33	160	210	Mar. 64
Ariz:	Phoenix	1.14	1.20	1.7	1.8	5	<5	3	4	25	30	Jan. 64
Ark:	Little Rock	1.19	1.20	1.3	1.5	105	10	44	41	170	140	Mar. 64
Calif:	Sacramento	1.18	1.24	1.6	1.6	10	<5	9	9	50	70	Dec. 63
	San Francisco	1.17	1.18	1.6	1.6	20	5	9	9	65	70	Feb. 64
C. Z:	Cristobal		1.15		1.5		<5		5		40	b
Colo:	Denver	1.18	1.22	1.6	1.6	40	10	22	18	110	85	Dec. 63
Conn:	Hartford	1.16	1.21	1.6	1.7	40	10	31	24	215	165	Dec. 63
Del:	Wilmington	1.18	1.23	1.6	1.6	35	5	30	24	150	120	Jan. 64
D. C:	Washington	1.13	1.17	1.5	1.5	45	10	21	18	110	85	Feb. 64
Fla:	Tampa	1.19	1.23	1.4	1.4	25	<5	14	17	275	255	Jan. 64
Ga:	Atlanta	1.18	1.18	1.4	1.5	75	10	33	29	190	120	Feb. 64
Hawaii:	Honolulu	1.18	1.18	1.6	1.7	15	5	12	10	85	80	Mar. 64
Idaho:	Idaho Falls	1.17	1.19	1.5	1.6	60	15	33	25	160	170	Jan. 64
Ill:	Chicago	1.21	1.19	1.6	1.7	30	5	23	22	125	110	Feb. 64
Ind:	Indianapolis	1.19	1.25	1.6	1.6	40	5	24	21	110	100	Dec. 63
Iowa:	Des Moines	1.18	1.22	1.5	1.5	50	10	28	25	85	100	Jan. 64
Kans:	Wichita	1.16	1.14	1.5	1.6	35	10	24	20	80	65	Mar. 64
Ky:	Louisville	1.14	1.16	1.4	1.5	100	10	39	32	125	100	Jan. 64
La:	New Orleans	1.21	1.23	1.4	1.5	80	15	41	46	160	140	Mar. 64
Maine:	Portland	1.18	1.23	1.6	1.7	55	10	38	36	300	240	Jan. 64
Md:	Baltimore	1.14	1.11	1.4	1.5	60	10	25	19	145	95	Jan. 64
Mass:	Boston	1.18	1.25	1.6	1.7	70	10	47	38	320	255	Feb. 64
Mich:	Detroit	1.17	1.21	1.6	1.6	30	5	22	20	130	125	Dec. 63
	Grand Rapids	1.18	1.23	1.6	1.6	35	5	22	22	145	130	Jan. 64
Minn:	Minneapolis	1.17	1.16	1.5	1.7	70	20	34	36	170	155	Feb. 64
Miss:	Jackson	1.22	1.28	1.4	1.4	80	10	36	36	125	100	Dec. 63
Mo:	Kansas City	1.15	1.21	1.6	1.5	55	15	29	30	75	85	Dec. 63
	St. Louis	1.16	1.23	1.5	1.6	45	10	25	25	85	90	Mar. 64
Mont:	Helena	1.16	1.23	1.4	1.5	80	15	39	26	245	200	Mar. 64
Nebr:	Omaha	1.17	1.24	1.6	1.5	50	15	29	28	100	95	Jan. 64
Nev:	Las Vegas	1.16	1.04	1.6	1.7	20	5	12	11	90	75	Feb. 64
N. H:	Manchester	1.20	1.22	1.6	1.7	60	10	43	37	355	280	Jan. 64
N. J:	Trenton	1.16	1.22	1.6	1.7	30	5	26	18	160	115	Dec. 63
N. Mex:	Albuquerque	1.12	1.14	1.6	1.5	20	5	12	9	50	55	Mar. 64
N. Y:	Buffalo	1.16	1.18	1.7	1.7	40	10	26	21	170	150	Dec. 63
	New York	1.14	1.21	1.6	1.7	60	15	41	31	225	175	Feb. 64
	Syracuse	1.19	1.20	1.6	1.7	45	5	31	22	165	145	Mar. 64
N. C:	Charlotte	1.17	1.19	1.4	1.5	80	10	35	30	155	110	Mar. 64
N. Dak:	Minot	1.16	1.19	1.5	1.6	130	20	60	50	170	140	Jan. 64
Ohio:	Cincinnati	1.20	1.25	1.6	1.6	40	10	28	24	95	90	Dec. 63
	Cleveland	1.16	1.22	1.6	1.7	40	<5	24	22	125	110	Mar. 64
Okla:	Oklahoma City	1.13	1.16	1.4	1.5	55	5	22	21	90	70	Feb. 64
Ore:	Portland	1.22	1.26	1.4	1.7	50	20	33	36	190	205	Dec. 63
Pa:	Philadelphia	1.15	1.22	1.6	1.6	35	<5	26	21	140	110	Mar. 64
	Pittsburgh	1.20	1.25	1.6	1.6	60	5	36	28	190	145	Mar. 64
P. R:	San Juan	1.14	1.17	1.4	1.6	35	<5	14	12	95	65	Mar. 64
R. I:	Providence	1.19	1.20	1.7	1.6	45	5	35	26	225	170	Jan. 64
S. C:	Charleston	1.17	1.24	1.4	1.5	60	10	26	27	150	125	Dec. 63
S. Dak:	Rapid City	1.07	1.27	1.5	1.8	100	25	49	46	175	180	Feb. 64
Tenn:	Chattanooga	1.20	1.24	1.4	1.5	110	15	46	36	185	115	Feb. 64
	Memphis	1.18	1.23	1.4	1.5	75	10	33	27	100	85	Jan. 64
Tex:	Austin	1.14	1.16	1.5	1.6	20	<5	10	8	55	50	Feb. 64
	Dallas	1.16	1.20	1.5	1.5	40	<5	19	19	75	80	Dec. 63
Utah:	Salt Lake City	1.13	1.17	1.5	1.6	55	20	30	33	205	160	Mar. 64
Vt:	Burlington	1.16	1.22	1.6	1.6	55	10	39	29	245	200	Dec. 63
Va:	Norfolk	1.15	1.18	1.4	1.6	45	5	23	22	115	95	Dec. 63
Wash:	Seattle	1.20	1.24	1.5	1.6	55	10	36	27	205	160	Feb. 64
	Spokane	1.22	1.25	1.5	1.6	55	20	30	29	160	165	Dec. 63
W. Va:	Charleston	1.13	1.15	1.4	1.5	85	10	34	26	120	85	Feb. 64
Wis:	Milwaukee	1.24	1.29	1.8	1.8	30	5	21	20	130	135	Jan. 64
Wyo:	Laramie	1.15	1.19	1.5	1.6	45	10	27	23	125	110	Jan. 64
Network average		1.17	1.21	1.5	1.6	52	9	28.4	24.8	147	125	Nov. 63

\* The monthly average iodine-131 and barium-140 concentration at each station was &lt;10 pc/liter.

b Station began operation in October 1963.

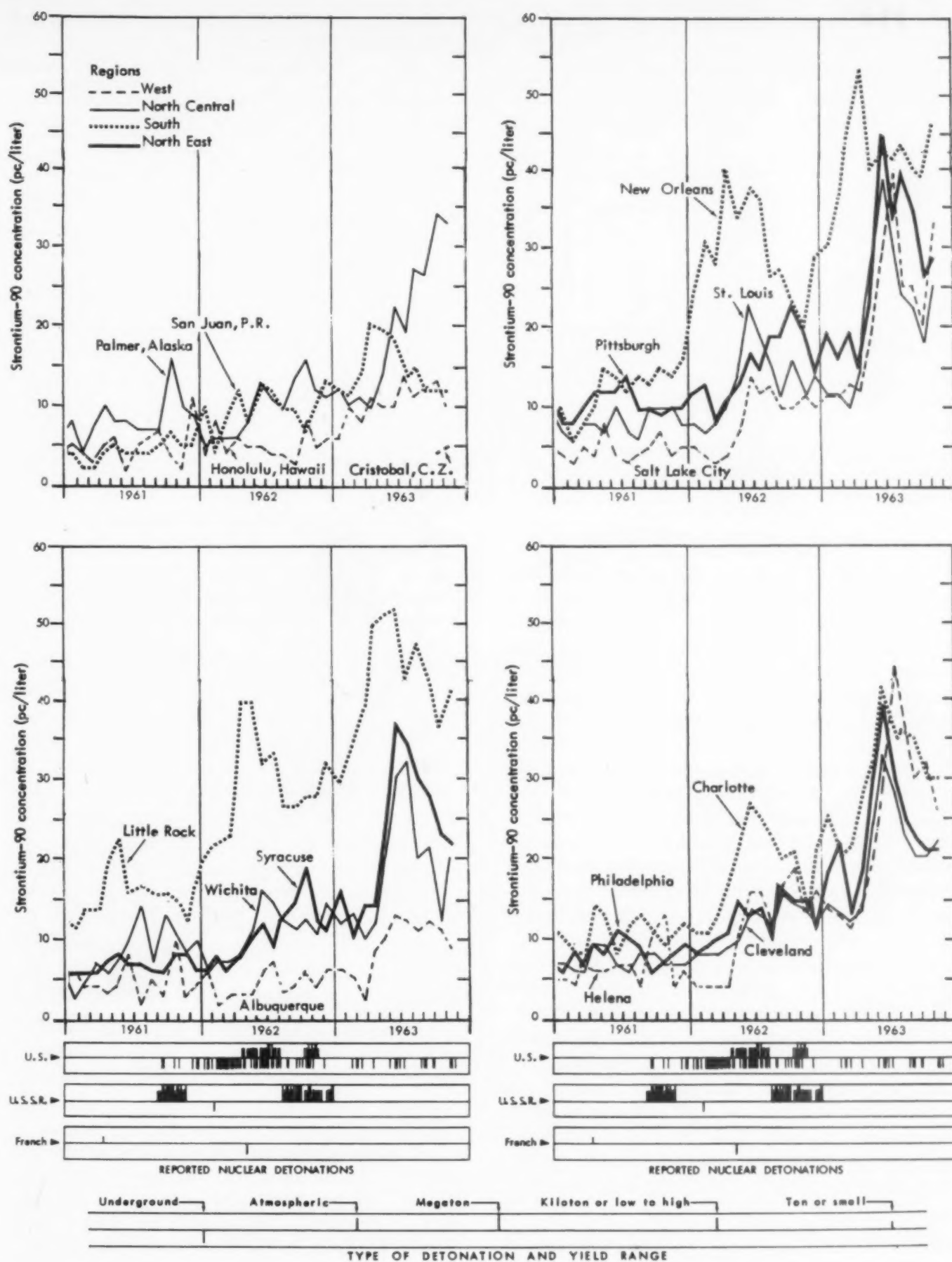


FIGURE 4.—STRONTIUM-90 CONCENTRATIONS IN PASTEURIZED MILK

TABLE 3.—DISTRIBUTION OF SAMPLING STATIONS IN VARIOUS RANGES OF RADIONUCLIDE CONCENTRATIONS IN MILK, NOVEMBER 1963

Strontium-89		Strontium-90		Iodine-131		Cesium-137		Barium-140	
Range (pc/liter)	Number of stations	Range (pc/liter)	Number of stations	Range (pc/liter)	Number of stations	Range (pc/liter)	Number of stations	Range (pc/liter)	Number of stations
<5-5	26	<1-4	1	<10	63	<5-45	2	<10	63
10	24	5-9	5			50-95	21		
15	7	10-14	3			100-145	23		
20	5	15-19	6			150-195	9		
25	1	20-24	17			200-245	5		
		25-29	14			250-295	3		
		30-34	6						
		35-39	7						
		40-50	4						

## 2. California Milk Network,<sup>2</sup> July-September 1963

State of California,  
Department of Public Health

Surveillance of specific radionuclides in milk is one phase of California's Department of Public Health program of radiation control. This milk monitoring function has been conducted at 8 milksheds since January 1960 by the Department's Bureau of Radiological Health, a constituent of the Division of Environmental Sanitation. Since the addition of the Del Norte and Mendocino milksheds to the program in March 1962, sampling of milk weekly or biweekly has been conducted at 10 major milksheds (see figure 5). The original sampling locations were chosen by the State Department of Agriculture as being representative of milk consumed by a high percentage of the population of the State.

Radiostrontium is separated chemically and counted in a low background counter, usually for a 60-minute period. Potassium-40, iodine-131, cesium-137, and barium-140 in fluid milk are determined by gamma scintillation spectroscopy using a sodium iodide crystal. A normal counting time of 100 minutes is used. The stable potassium content of milk (g/liter) may be estimated by multiplying the potassium-40 concentration (pc/liter) by  $1.18 \times 10^{-3}$ .

<sup>2</sup> Data from *Radiological Health News*, Vol. 3, No. 1, Bureau of Radiological Health, State of California Department of Public Health, 2151 Berkeley Way, Berkeley 4, California.



FIGURE 5.—CALIFORNIA MILKSHEDS

## Results

The monthly averages of the radionuclide and calcium data for milk for the period July-September 1963 are presented in table 4. A summary of the 1961-1963 results by Heslep and Cornish appears in the December 1963 *Radiological Health Data* (2).

TABLE 4.—RADIONUCLIDES IN CALIFORNIA MILK, JULY-SEPTEMBER 1963\*

[Radioactivity concentrations in pc/liter]

Element and month	Del Norte	Fresno	Humboldt	Los Angeles	Mendocino	Sacramento	San Diego	Santa Clara	Shasta	Sonoma	Average
Calcium (g/liter)											
July.....	1.31	1.18	1.27	1.09	1.21	1.14	1.13	1.14	1.16	1.18	1.18
August.....	1.25	1.16	1.21	1.10	1.20	1.15	1.14	1.12	1.14	1.16	1.16
September.....	1.29	1.13	1.40	1.12	1.20	1.20	1.12	1.20	1.23	1.23	1.21
Potassium-40											
July.....	1195	1260	1158	1301	1295	1249	1255	1294	1311	1267	1249
August.....	1170	1265	1094	1315	1285	1204	1280	1204	1208	1232	1226
September.....	1145	1257	1136	1235	1300	1220	1244	1293	1274	1280	1238
Strontium-89											
July.....	141	18	33	11	22	22	8	24	40	26	35
August.....	108	10	19	7	19	10	3	19	21	21	24
September.....	43	6	11	5	10	6	3	7	14	11	12
Strontium-90											
July.....	57.4	7.6	12.8	4.1	8.4	8.3	4.0	8.6	17.0	11.0	13.9
August.....	43.6	6.1	14.1	6.2	9.3	7.2	4.4	8.1	11.5	9.5	12.0
September.....	45.8	5.1	12.3	5.6	9.9	8.3	3.5	5.4	14.0	10.3	12.0
Cesium-137											
July.....	179	72	56	45	38	57	38	86	92	70	73
August.....	140	53	79	47	52	40	27	87	69	54	65
September.....	131	53	60	50	33	43	27	55	71	59	58

\* The monthly iodine-131 and barium-140 averages at all stations during this period were zero.

Note: All values reported are the best available estimate. If the counting rate of the sample is not equal to at least twice the 95-percent error, a "b" would be printed beside the value.

### 3. Florida Milk Network, November 1962-December 1963

*Division of Radiological and Occupational  
Health, Florida State Board of Health*

The Florida State Board of Health began sampling raw milk for iodine-131 analysis in two major areas of the State in November 1962. The program has since been expanded to include the sampling of milk from five areas shown in figure 6. Radiostrontium and cesium-37 analyses are also conducted but results are not available for publication. A Regional State Board of Health Laboratory is located in each of these areas. Milk samples are sent first to the regional laboratories where the samples are passed through ion exchange columns. The column resins are removed and sent to the State Radiological Health Laboratory in Orlando for analysis.

Milk produced in the counties comprising each area is generally processed, marketed and consumed in that area. These areas are characterized by differences in dairying practices related to the gradual transition from small farms using locally-grown feed in the "West Florida" region to larger farms using different types of grass and predominantly purchased feeds in the southern areas.

March 1964



FIGURE 6.—FLORIDA MILK SAMPLING LOCATIONS

#### Sampling

Raw milk samples are taken from a tank truck, the route of which passes by farms which are widely dispersed over the area represented. Where there is no route representative of a large portion of the area, samples are collected directly from selected farms and composited. Samples were collected weekly when iodine-131 was detectable in milk. Presently the sampling is on a monthly basis.

## Analytical Procedures

Iodine-131 analyses are performed by using an ion exchange technique. From November 1962 through July 1963, one-gallon samples were run through 80 ml of resin and counted with a 2" x 2" sodium iodide crystal and single channel analyzer. Sensitivity was increased in August and September 1963 by use of a 4" x 4" crystal and multi-channel analyzer. Improvements in analytical methods instituted in October 1963 have permitted the same sensitivity to be obtained using only one-liter samples. These samples are passed through 15 ml of resin. The resin is then mounted in a 3" x 3" well crystal connected to a single channel analyzer.

The monthly averages of iodine-131 con-

TABLE 5.—IODINE-131 IN FLORIDA RAW MILK  
[Average concentrations in pc/liter]

Month	Sampling location				
	West Florida	N. E. Florida	Central Florida	Tampa Bay Area	S. E. Florida
Nov. 62	147	53	*	—	—
Dec. 62	93	69	—	—	—
Jan. 63	65	52	—	—	—
Feb. 63	14	9	11	—	—
Mar. 63	3	<2	<2	—	—
Apr. 63	<2	<2	<2	—	—
May 63	<2	<2	<2	—	—
June 63	<2	<2	<2	—	—
July 63	<2	<2	<2	<2	<2
Aug. 63	<2	<2	<2	<2	<2
Sep. 63	<2	<2	<2	<2	<2
Oct. 63	<2	<2	<2	<2	<2
Nov. 63	<2	<2	<2	<2	<2
Dec. 63	<2	<2	<2	<2	<2

\* A dash indicates sampling not begun.

centrations in milk produced in Florida are shown in table 5.

## 4. Indiana Milk Network, November 1963

Bureau of Environmental Sanitation,  
Indiana State Board of Health

The Indiana State Board of Health began sampling pasteurized milk for radiological analysis in September 1961. Indiana was geographically divided into five major milksheds, and one large dairy within each milkshed was selected as a sampling section (see figure 7).

The milk samples are routinely analyzed for iodine-131, cesium-137, barium-140, strontium-89 and strontium-90. Until August 1963, analyses for the gamma emitters iodine-131, cesium-137 and barium-140 were conducted on a weekly basis, except when iodine-131 exceeded 100 pc/liter, at which times the frequency of sampling was increased. Because of continued low concentrations of the short-half-lived gamma emitters, the sampling frequency was reduced in August 1963 to once per month for the northeast, southeast and southwest milksheds. Strontium-89 and strontium-90 analyses are performed monthly for each station.

An ion exchange analytical procedure (3) is used for strontium-89 and strontium-90 analyses. A 512-channel pulse height analyzer and shielded 4 x 4-inch sodium iodide crystal are used for the gamma analysis of iodine-131, cesium-137, and barium-140.

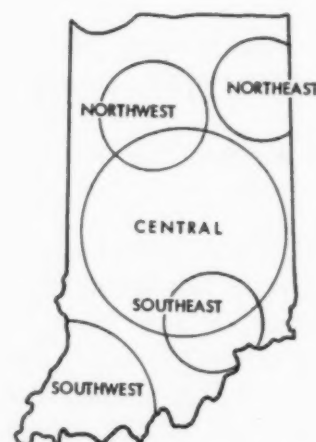


FIGURE 7.—INDIANA MILK SAMPLING LOCATIONS

The monthly averages of the data obtained for the individual sampling stations and the State averages are reported in table 6. The State average is an arithmetic average of the station values.

TABLE 6.—RADIONUCLIDES IN INDIANA MILK,  
NOVEMBER 1963

[Concentrations in pc/liter]

Sampling location	Sr <sup>89</sup>	Sr <sup>90</sup>	I <sup>131</sup>	Cs <sup>137</sup>	Ba <sup>140</sup>
Northeast.....	15	15	<10	120	<10
Southeast.....	15	19	<10	110	<10
Central.....	5	23	<10	95	<10
Southwest.....	15	16	<10	80	<10
Northwest.....	20	15	<10	115	<10
State average...	14	18	<10	104	<10

## 5. Pennsylvania Milk Network, September 1962-November 1963

*Bureau of Environmental Health,  
Pennsylvania Department of Health*

Samples of pasteurized milk are routinely collected from 10 major milk consumption areas throughout Pennsylvania (see figure 8). Two samples per week are collected in Philadelphia and Pittsburgh, while weekly composite samples are collected from the other eight stations. At each sampling location subsamples are collected from the major dairies, which supply the area and are composited in proportion to the amount of milk processed by each dairy. This composite sample is then sent to the Radiation Laboratory of the Division of Occupational Health in Harrisburg for analysis. Iodine-131 analyses were carried out from September 1962 through January 1963 when concentrations fell to nondetectable levels. Analysis for strontium-90 is carried out on all samples. The strontium-90 program was initiated on a state-wide basis in April 1963.

The chemical separation technique used for strontium-90 is essentially an ion exchange method described by Porter, *et al.* (3) One liter of milk is passed through an ion exchange column. Yttrium-90 is eluted from the resin and is counted in an automatic low background proportional counter with a window thickness of 0.8 mg/cm<sup>2</sup>. Iodine was also separated by passing one liter of milk through an ion ex-



FIGURE 8.—PENNSYLVANIA MILK SAMPLING STATIONS AND MILK CONSUMPTION AREAS

change column (4). Iodine-131 counting was performed with 2" x 2" sodium iodide gamma scintillation crystal and multichannel analyzer.

The monthly average strontium-90 levels in pasteurized milk are shown in table 7 and the monthly average iodine-131 results are shown in table 8.

TABLE 8.—IODINE-131 IN PENNSYLVANIA MILK, SEPTEMBER 1962-JANUARY 1963

[Concentrations in pc/liter]

Sampling location	September	October	November	December	January
Dauphin.....	40	80	80	20	<10
Erie.....	80	110	60	20	<10
Kingston.....	40	90	95	35	<10
Lancaster.....	20	50	45	35	<10
Pittsburgh.....	60	110	200	30	<10
Reading.....	30	60	70	20	<10
Williamsport.....	40	70	55	25	<10
York.....	20	70	40	10	<10
State average...	41.2	80	80.6	24.4	<10

TABLE 7.—STRONTIUM-90 IN PENNSYLVANIA MILK, APRIL-NOVEMBER 1963

[Concentrations in pc/liter]

Sampling location	April	May	June	July	August	September	October	November
Altoona.....	12	*	18	50	41	38	33	8
Dauphin.....	7	16	33	29	35	25	22	18
Erie.....	25	20	40	42	41	29	34	20
Kingston.....	15	10	25	42	48	36	30	25
Lancaster.....	7	24	22	22	28	38	25	13
Philadelphia.....	21	28	29	22	31	31	25	—
Pittsburgh.....	15	20	26	44	54	22	20	12
Reading.....	12	21	22	36	43	27	21	14
Williamsport.....	9	14	18	36	41	49	24	11
York.....	12	24	18	32	24	20	20	12
State Average.....	13.5	19.7	25.1	35.5	38.6	31.5	25.4	14.7

\* No sample.

## 6. Canadian Milk Network,<sup>3</sup> November 1963

*Radiation Protection Division,  
Department of National Health and Welfare,  
Ottawa, Canada*

The Radiation Protection Division of the Department of National Health and Welfare began monitoring milk for strontium-90 in November 1955. At first, analyses were carried out on samples of powdered milk obtained from processing plants. However, since January 1963, liquid whole milk has been analyzed instead. With this change, more representative samples of milk consumed can be obtained, and in addition it is possible to choose milk sampling locations (see figure 9) in the same areas as the air and precipitation stations. At present, the analyses include determinations of iodine-131, strontium-89, cesium-137, and strontium-90 as well as stable potassium and calcium.

The milk samples are obtained through the cooperation of the Marketing Division of the Canadian Department of Agriculture. At each station samples are collected three times a week from selected dairies, combined into weekly composites, and forwarded to the radiochemical laboratory in Ottawa. The contribution of each dairy to the composite sample is directly proportional to its volume of sales. In most cases a complete sample represents over 80 percent of the milk processed and distributed in the

<sup>3</sup>Data from *Radiation Protection Programs, Vol. 1, No. 12*: 25-30, Radiation Protection Division, Canadian Department of National Health and Welfare. (December 1963)



FIGURE 9.—CANADIAN MILK SAMPLING STATIONS

area. Several of the weekly samples are randomly selected and analyzed for iodine-131. The results of the spot checks for iodine-131 will not be reported unless there is evidence that the levels are rising. A monthly composite of the samples is analyzed for strontium-90, strontium-89, cesium-137, and stable potassium and calcium.

### *Analytical Methods*

For the analysis of iodine-131, radiochemical methods are used (5). Carrier iodine is added and the milk is then evaporated in the presence of sodium hydroxide and ashed. The iodine ion is oxidized to free iodine and extracted with carbon tetrachloride, back-extracted in sulfite solution, and precipitated as silver iodide. The precipitate is counted in a low background beta counter and the iodine-131 determined by comparison with standard preparations.

For the analysis of radiostrontium, carrier strontium is added to one-liter sample of milk, and the milk is then evaporated under infra-red lamps in a tray lined with a polyethylene sheet. The residue is ashed in a muffle furnace at 450°C, dissolved in dilute nitric acid, and strontium is separated by burning nitric acid precipitations. The combined strontium-89 and strontium-90 are determined by counting in a low background beta counter. Strontium-90 is determined separately by extracting and counting the yttrium-90 daughter nuclide while strontium-89 is estimated by difference from the total radiostrontium measurement. Appropriate corrections are made for self-absorption and counter efficiency at all stages. Calcium is determined by flame photometry.

Cesium-137 is determined by gamma spectrometry using a scintillation crystal and a multi-channel pulse height analyzer. A sample consisting of 4.5 liters of milk is placed in a sample tray constructed in the form of an inverted well to accommodate the 5 x 4-inch sodium iodide crystal detector. The sample is counted for 100 minutes and the gamma spectrum recorded. Estimates are made of the potassium-40 and cesium-137 content of the milk by comparison of the spectrum with standard preparations. Stable potassium content is estimated from the potassium-40 concentrations.

## Sources of Error

In the iodine and strontium determinations, tests indicate that the statistical error (95 percent confidence level) in the chemical operations involved is about plus-or-minus 10 percent. This value is independent of the concentration of the radioisotope in the milk because it depends only on the recovery of the "carrier." In the determination of cesium this factor is not involved.

The operational error must be combined with the counting error, which depends primarily on the concentration of the nuclide in the sample, the background radiation, and the length of time the sample and background are counted. This counting error has been evaluated mathematically for the particular counting arrangement used.

The overall errors, estimated on the basis indicated above, were previously published (6). For example, the  $2\sigma$  total errors (representing 95 percent confidence) associated with a measured concentration of 10 pc/liter, in units of pc/liter are  $\text{Sr}^{89}$ , 2.5;  $\text{Sr}^{90}$ , 1.5;  $\text{I}^{131}$ , 5; and  $\text{Cs}^{137}$ , 6.

## Results

Table 9 presents monthly averages of strontium-89, strontium-90, cesium-137, and stable calcium and potassium in Canadian whole milk. Spot checks for iodine-131 indicate that all samples had  $<5$  pc/liter. Figure 10 shows the variation of the network average radionuclide concentrations of Canadian whole milk.

TABLE 9.—RADIONUCLIDES IN CANADIAN WHOLE MILK, NOVEMBER 1963

(Radionuclide concentrations in pc/liter)

Station	Calcium (g/liter)	Potassium (g/liter)	Strontium-89	Strontium-90	Cesium-137
Calgary.....	1.05	1.5	24	50.4	298
Edmonton.....	1.25	1.4	14	33.6	194
Ft. William.....	1.29	1.4	17	57.5	251
Fredericton.....	1.25	1.4	20	56.5	338
Halifax.....	1.26	1.5	14	43.0	271
Montreal.....	1.26	1.5	17	38.6	231
Ottawa.....	1.26	1.5	15	29.3	192
Quebec.....	1.23	1.6	20	55.5	328
Regina.....	1.19	1.5	25	51.5	174
St. John's Nfld.....	1.25	1.3	15	51.2	230
Saskatoon.....	1.25	1.3	21	46.7	172
Sault Ste. Marie.....	1.21	1.5	22	41.3	219
Toronto.....	1.26	1.5	7	19.5	127
Vancouver.....	1.38	1.4	23	44.5	378
Windsor.....	1.22	1.5	6	17.2	81
Winnipeg.....	1.34	1.4	22	41.0	209
Average.....	1.25	1.4	17	42.3	230

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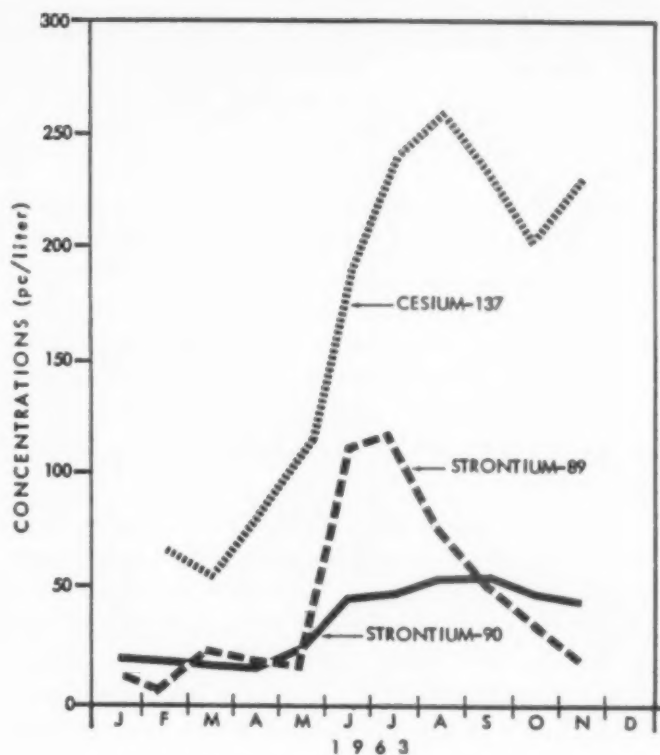


FIGURE 10.—NETWORK AVERAGE STRONTIUM-89, STRONTIUM-90 AND CESIUM-137 CONCENTRATIONS IN CANADIAN WHOLE MILK

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# MOVING ANNUAL AVERAGE RADIONUCLIDE CONCENTRATIONS IN PASTEURIZED MILK, DECEMBER 1962-NOVEMBER 1963

*Division of Radiological Health,  
Public Health Service*

Radionuclide concentration values reported by the Pasteurized Milk Network (1) can be used to assess the contribution of milk to an individual's or a population's radiation exposure. This is done by determining both the annual average concentrations of specific radionuclides in milk and the average daily milk consumption of an individual or a suitable sample of the population.

The data listed in table 1 are concerned with the first of these requirements, *i.e.*, annual average concentrations of strontium-89, strontium-90, iodine-131 and cesium-137 in one liter of pasteurized milk. Limited data are available for estimating the average daily milk consumption (on a volume basis) for specific age groups in the U. S. population (2, 3).

To arrive at a basis of comparison between the daily rates of intake of the radionuclides from the milk component of the diet and the Federal Radiation Council's ranges of transient daily rates of intake (4), it is assumed that the average daily milk consumption of an individual in a population group is one liter. The Guides, however, apply to total intake from all foods. The upper limits of Range II correspond to the Radiation Protection Guide (RPG) for iodine-131 and one-third of the Radiation Protection Guide for radioactive strontium. The Guides are, for administrative reasons, expressed as a yearly radiation dose, but are based on lifetime exposure (5). However, the FRC emphasizes that the annual acceptable risk or exposure dose is not a dividing line between safety and danger in actual radiation situations (6). The ICRP has set the maximum permissible concentration (MPC) for cesium-137 in water for the population at large equal to 2000 pc/liter (7). This MPC may be applied to milk, if it is assumed that all food would be contaminated to the same extent.

Annual averages of radionuclide concentrations in milk sampled by the PHS Pasteurized Milk Network are presented in table 1. The

data in table 1 are calculated as follows: (a) results from all samples collected in each week (Sunday through Saturday) are averaged, (b) the averages for all weeks ending in twelve consecutive months are averaged to obtain the annual average.<sup>1</sup> To obtain the annual average daily intake (pc/day) of radionuclides from milk, the annual average concentration values (pc/liter) in table 1 must be multiplied by the annual average daily consumption (liters/day) of milk (3, 4).

Monthly variations of radionuclide concentrations in milk are due to a number of combined causes. The moving yearly average (table 1), obtained by updating the previous twelve-month average by one month, shows variations averaged over the year and tends to minimize purely seasonal variations. This method, therefore, shows trends over a considerable period of time.

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<sup>1</sup> Beginning with the October 1963 data, iodine-131 values of <10 pc/liter are considered to be zero for averaging purposes; previously 5 pc/liter was used in calculating the average.

TABLE 1.—MOVING ANNUAL AVERAGE RADIONUCLIDE CONCENTRATIONS IN MILK\*

[Concentrations in pc/liter]

Sampling locations		Strontium-89		Strontium-90		Iodine-131		Cesium-137	
		November 1962– October 1963	December 1962– November 1963	November 1962– October 1963	December 1962– November 1963	November 1962– October 1963	December 1962– November 1963	November 1962– October 1963	December 1962– November 1963
Ala:	Montgomery.....	67	64	20	20	14	9	68	72
Alaska:	Palmer.....	37	33	17	19	11	8	95	108
Ariz:	Phoenix.....	15	14	4	4	8	6	20	22
Ark:	Little Rock.....	135	125	40	41	32	24	138	143
Calif:	Sacramento.....	42	39	9	9	9	7	51	55
	San Francisco.....	75	73	12	13	9	8	61	65
Colo:	Denver.....	27	26	15	16	8	7	78	81
Conn:	Hartford.....	26	23	20	21	9	5	125	133
Del:	Wilmington.....	36	30	23	24	19	8	111	115
D. C:	Washington.....	43	40	19	19	13	7	87	89
Fla:	Tampa.....	36	33	13	14	17	11	196	206
Ga:	Atlanta.....	93	89	27	28	20	14	126	131
Hawaii:	Honolulu.....	37	36	10	10	11	9	66	69
Idaho:	Idaho Falls.....	45	43	20	22	11	7	112	121
Ill:	Chicago.....	24	21	18	19	10	6	92	96
Ind:	Indianapolis.....	35	31	21	21	16	9	84	88
Iowa:	Des Moines.....	66	60	22	23	22	12	78	82
Kana:	Wichita.....	48	44	17	18	19	12	67	69
Ky:	Louisville.....	101	91	30	31	20	9	93	97
La:	New Orleans.....	147	141	38	40	24	19	136	142
Maine:	Portland.....	31	26	27	28	11	5	169	179
Md:	Baltimore.....	51	48	20	20	13	7	107	109
Mass:	Boston.....	36	32	30	31	8	5	180	192
Mich:	Detroit.....	21	17	19	19	12	6	96	101
	Grand Rapids.....	22	19	18	19	11	7	99	104
Minn:	Minneapolis.....	53	49	26	27	13	7	130	135
Miss:	Jackson.....	147	139	32	34	23	17	98	102
Mo:	Kansas City.....	80	73	24	25	27	16	71	74
	St. Louis.....	54	50	20	21	14	8	74	77
Mont:	Helena.....	51	48	24	25	19	14	139	149
Nebr:	Omaha.....	56	51	23	24	23	16	85	89
Nev:	Las Vegas.....	19	19	7	8	6	5	59	61
N. H:	Manchester.....	33	29	28	30	9	6	202	215
N. J:	Trenton.....	26	22	20	20	13	6	103	107
N. Mex:	Albuquerque.....	24	23	8	9	10	8	36	39
N. Y:	Buffalo.....	24	22	20	21	6	5	116	121
	New York.....	34	30	25	27	12	6	127	135
	Syracuse.....	27	25	21	22	8	5	107	113
N. C:	Charlotte.....	72	67	29	30	8	6	104	108
N. Dak:	Minot.....	85	81	42	44	10	5	118	123
Ohio:	Cincinnati.....	45	39	24	24	21	10	76	79
	Cleveland.....	29	25	19	20	14	7	91	95
Okla:	Oklahoma City.....	75	69	22	22	29	21	80	82
Ore:	Portland.....	98	83	26	27	15	9	131	143
Pa:	Philadelphia.....	33	28	22	22	17	7	105	108
	Pittsburgh.....	41	36	26	26	22	9	125	130
P. R:	San Juan.....	* 87	79	* 14	14	* 15	14	* 89	87
R. I:	Providence.....	31	26	24	25	12	6	135	142
S. C:	Charleston.....	73	69	25	26	16	13	108	114
S. Dak:	Rapid City.....	70	67	30	33	16	9	121	131
Tenn:	Chattanooga.....	118	110	33	35	17	10	122	127
	Memphis.....	103	96	29	30	22	14	78	81
Tex:	Austin.....	39	36	9	9	30	24	42	45
	Dallas.....	90	85	20	20	45	38	74	77
Utah:	Salt Lake City.....	36	34	19	20	10	8	127	135
Vt:	Burlington.....	31	27	24	25	10	4	140	149
Va:	Norfolk.....	51	47	21	22	12	7	92	94
Wash:	Seattle.....	71	59	23	24	15	8	135	141
	Spokane.....	53	50	23	24	10	7	117	125
W. Va:	Charleston.....	72	66	28	28	11	6	87	89
Wis:	Milwaukee.....	24	20	16	17	14	6	94	99
Wyo:	Laramie.....	43	40	19	20	5	5	109	113
Network average.....		55	50	24.8	22.7	15	10	103	108

\* Annual averages were computed on basis of 52 weekly averages.

b Annual averages were computed on basis of 53 weekly averages.

c Average is for 48 weeks (No sample was received in November 1962).

# THE ANALYTICAL QUALITY CONTROL SERVICE OF THE DIVISION OF RADIOLOGICAL HEALTH—INTERLABORATORY STUDY OF IODINE-131 SURVEILLANCE MEASUREMENTS IN MILK

Marvin Rosenstein and Abraham S. Goldin<sup>1</sup>

Ever since the Division of Radiological Health (DRH) began to measure levels of radioactivity in environmental samples, quality control of these measurements has been emphasized. Since October 1962, the quality control program has been placed on a more formal basis, under the designation of Analytical Quality Control Service, DRH. In this report the objectives and operations of this program are reviewed, along with the fundamental means of meeting these objectives. Included also as an example of the functioning of the program is a summary of the fields of investigation and the data from the first quality control study on iodine-131 in milk.

## *Structure of the Analytical Quality Control Service (AQCS)*

The Analytical Quality Control Service consists of three general activities: intralaboratory quality control, interlaboratory quality control, and outside assistance. *Intralaboratory* quality control has as its objective the maintenance of precision in analyses, and is basically the responsibility of the individual DRH laboratories. The AQCS serves, however, as a general coordinating group in establishing criteria for the intralaboratory programs. *Interlaboratory* quality control establishes the overall validity of results originating from the several DRH laboratories and endeavors to assist in improving analytical conditions that are not acceptable. Outside assistance is provided to State and local radiological health laboratories and DRH contract laboratories.

## *Objectives and Mechanics of AQCS*

The objectives of AQCS are threefold. First, it maintains a continuous collection of representative radiological data for statistical analy-

sis as described below. Second, it determines the accuracy of these data. Third, it recommends steps to correct laboratory operations that do not meet the required limits of accuracy.

*Collection of Data*—Continuous collection of data is maintained through the distribution of appropriate environmental samples and the performance of technical experiments. The unknown environmental samples are analyzed by a round-robin procedure to measure the agreement among several laboratories. The technical experiments involve the analysis of (1) accurately known preparations (to obtain bias of results from a true value), and (2) blank preparations (to study the sensitivity of techniques and contamination of the laboratory). Each of these types of analysis has a different relationship to the determination of whether or not data are satisfactory.

*Determination of Accuracy*—Accuracy is determined by comparing statistically the agreement between observed data and correct values with expected or standard limits of variability. Selection of the proper limits of variability is of fundamental importance. These limits are chosen to reflect both the requirements for analytical measurements and laboratory capabilities under normal operating conditions. Once these limits have been chosen, conventional statistical tests can be applied to obtain the desired information concerning the accuracy of the observed data.

*Corrective Measures*—When corrective measures are indicated by the analysis of the data, two steps are taken to assure that valid decisions are made. First, using knowledge of the related disciplines involved in the radiological measurements, along with the statistical indications of the data, factors that could contribute to the type of inaccuracy observed are listed and given further consideration and study. Second, suggestions as to the possible causes of unacceptable results are forwarded to the laboratory submitting the results as recommendations for implementing the im-

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provement of the measurements. The laboratory, in turn, tests these recommendations and accepts those which prove to be valid. Subsequent samples are distributed until the process under study is brought under control. After this has been accomplished, periodic samples are issued to monitor laboratory operations.

### *Fields of Investigation*

The AQCS is responsible for providing quality control measures for all types of environmental radiological measurements. At present, it is focusing most attention on the results of Pasteurized Milk Network samples. An extensive study is being made on all the elements for which milk analyses are designed. Approximately 15-20 percent of the analytical workload is devoted to internal or external quality control measures. Iodine-131 and strontium-90 have been given particular emphasis because of their biological importance. Projects are underway for the study of analyses of diet samples, bone samples, and other environmental samples performed on a large scale.

Assistance to outside agencies has begun with the formation of a calibration standards program. This service makes available to these agencies nuclide preparations sufficient for calibrating instrumentation for radiological determinations. Plans are also in progress for sample distribution and quality control analyses similar to the DRH interlaboratory program.

### *Iodine-131 Quality Control Study*

Milk samples were analyzed for iodine-131 by gamma spectroscopy in the Division of Radiological Health Laboratories located in Las Vegas, Nevada; Montgomery, Alabama; and Winchester, Massachusetts. In addition to these which carry the bulk of the network analytical load, two other divisional laboratories which have gamma spectroscopy capabilities were included in the study. Five such laboratories were included in this study.

To evaluate the ability of these laboratories to obtain accurate results for this type of measurement, milk samples were prepared by adding a known amount of calibrated iodine-

131 solution to a master milk sample free of this nuclide. Evaporated milk which had been stored for several weeks met this requirement. In addition to the known amount of iodine-131, the milk samples contained the usual, but unknown concentrations of other radionuclide contaminants (cesium-137 and potassium-40). Subsamples of this preparation were distributed to the laboratories for analysis in accordance with a pre-planned schedule based on the control chart analysis (1) which was used in analyzing the resulting data. The data returned were then examined for accuracy; i.e., their agreement with the known iodine-131 value. The limits allowed for the variation of the observed iodine-131 values from the known iodine-131 values were calculated from an assigned standard deviation, based on past experiences of the capabilities of iodine-131 surveillance measurements and on a judgment as to the requirements for accuracy in this determination.

Two samples were distributed three weeks apart. Each milk sample was analyzed at three different time intervals, yielding a high level, an intermediate level and a low level. Figure 1 illustrates graphically as well as numerically the results of this study at the three iodine-131 concentrations. The improvement in the second sample results over those of the first sample results and their agreement with the expected values is apparent in figure 1. This figure expresses only the measurement of bias of the reported data. Precision within individual laboratories was also studied but is not discussed in this report. The analytical variations which are the subject of this report included the entire analytical system, from sample receipt at the laboratory through final calculations. The samples were utilized to detect data that were analytically unacceptable. It should be noted, however, that the discrepancies uncovered were not biologically significant.

To clarify the notations on the figure, the following explanations are given:

1. Expected Values: This is the amount of iodine-131 added to the milk and present at the time of analysis. These values are 460, 115 and 29 pc/liter.
2. Upper and Lower Warning Levels: This is the level between which the results are

expected to fall, based on knowledge of the analytical capabilities of the technique and overall accuracy requirements. These particular warning levels were

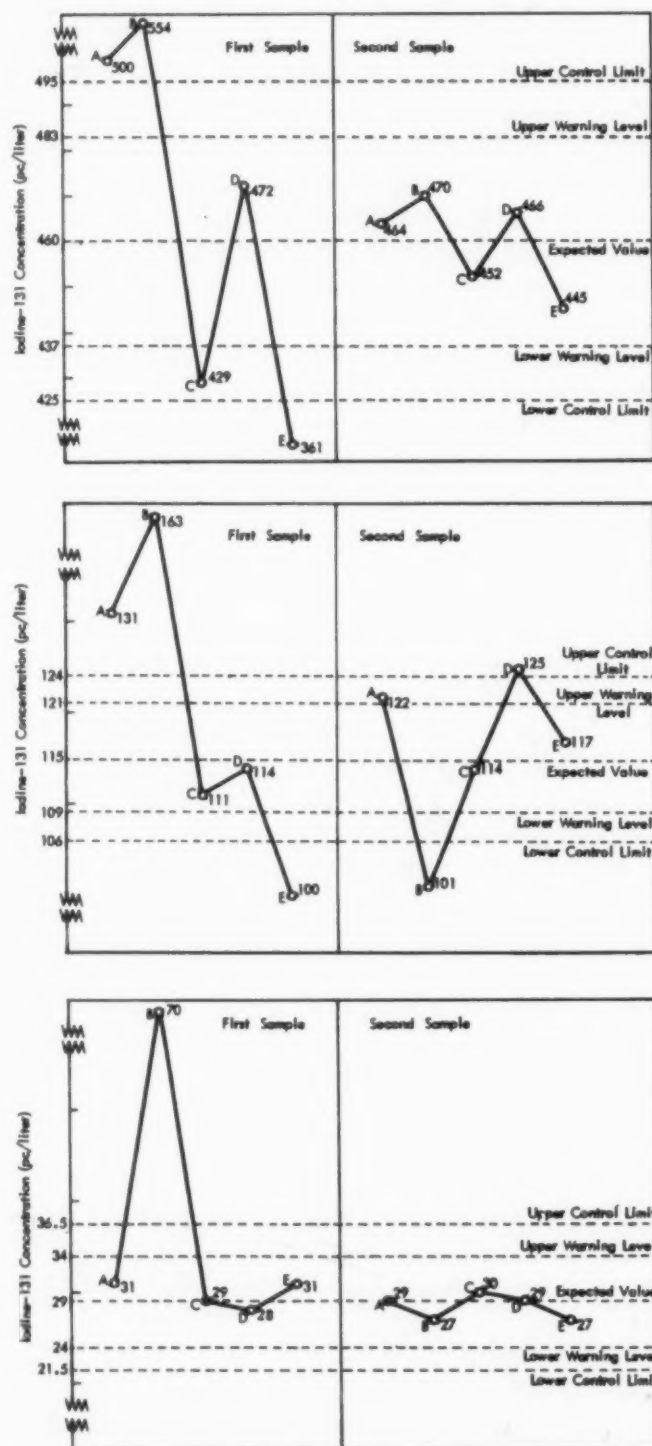


FIGURE 1.—IMPROVEMENT OF LABORATORY RESULTS OF IODINE-131 MEASUREMENTS IN MILK IN FIVE LABORATORIES (A, B, C, D, and E) FOR THREE CONCENTRATIONS OF ACTIVITY: 460, 115, and 29 pc/liter.

based on the selected overall variation for individual iodine-131 determinations of  $\pm 10$  pc/liter for levels  $< 100$  pc/liter and  $\pm 10$  percent for levels  $> 100$  pc/liter, at two standard deviations. Since each value given in figure 1 represents the mean of four determinations in each laboratory, the expected standard deviation of these means is obtained by dividing the standard deviation of individual determinations by the square root of four (the number of determinations from which the mean is calculated). Therefore, the mean values of the data have an expected overall variation of  $\pm 5$  pc/liter for levels  $\leq 100$  pc/liter and  $\pm 5$  percent for levels  $> 100$  pc/liter, at two standard deviations. These are the numbers used to describe the warning levels in figure 1. Any values falling within these levels are considered acceptable. Any values falling outside these levels warrant inspection.

- Upper and Lower Control Limits: These control limits were set at the  $\pm$  three standard deviation limits, which are, for means of four determinations,  $\pm 7.5$  pc/liter for  $\leq 100$  pc/liter levels and  $\pm 7.5$  percent for  $> 100$  pc/liter levels. Any values falling outside these limits indicate that corrective measures are necessary.
- A, B, C, D, E: These are the code names of the participating laboratories. Laboratories A, C, and D are the Division laboratories which routinely analyze milk by gamma spectroscopy.

From figure 1, one can see that the results on the first sample are in general out of control. After detailed investigation of the data, from which recommendations were made to correct several discrepancies, a period of time was then allowed for corrective measures to be taken. A second sample was then issued and the same analysis repeated. In figure 1, the second sample results show a marked improvement in the quality of iodine-131 data. These latter results therefore indicated that individual iodine-131 analyses are being performed within an accuracy of  $\pm 10$  percent at the two standard deviation level.

### Conclusions.

By the application of adequately designed samples, properly applied statistical methods, and appropriate corrective measures, the quality of radiological measurements can be maintained at an acceptable level. Thus, as in the case of the iodine-131 measurement study, once the analytical data appear to be under control, periodic checks can be made to continue surveillance and additional steps can be taken to improve the data even further.

The quality control procedure as applied to iodine-131 measurements in milk can be

adapted to any other nuclides in environmental samples provided that a master sample can be produced containing a known amount of the nuclide of interest and an acceptable level of variability can be established. For example the technique is presently being applied by the Analytical Quality Control Service to strontium-90, cesium-137, radium-226, potassium, and calcium analyses in various environmental samples.

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## RADIONUCLIDES IN INSTITUTIONAL DIET SAMPLES, JULY-SEPTEMBER 1963

*Division of Radiological Health, Public Health Service*

The determination of radionuclide concentrations in the diet constitutes an important element of an integrated program of environmental radiation surveillance and assessment. In recognition of the potential significance of the diet in contributing to total environmental radiation exposures, the Public Health Service (PHS) initiated its Institutional Diet Sampling Program in 1961. This program is administered by the Division of Radiological Health with the assistance of the Division of Environmental Engineering and Food Protection (1).

The program is designed to estimate the dietary intake of radionuclides in a selected population group ranging from children to young adults of school age. Initially, the program consisted of sampling diets in eight institutions, but it has since been expanded to 21 boarding schools or institutions, geographically distributed as shown in figure 1. Institutions selected range from financially well-to-do boarding schools to orphanages with severe economic

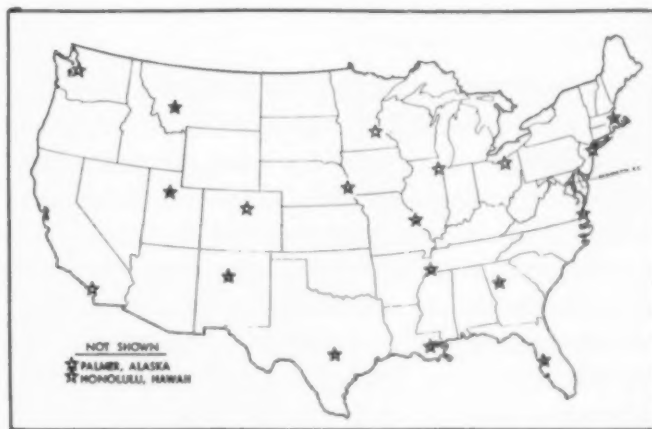


FIGURE 1.—INSTITUTIONAL DIET SAMPLING LOCATIONS

limitations. Each institution (sampling point) except the one at Los Angeles is located in a community from which the PHS Pasteurized Milk Network collects samples. The analytical data from this program supplement the findings for the Institutional Diet Sampling Program.

### *Sampling Procedure*

In general, the sampling procedure is the same at each institution. Each sample represents the edible portion of the diet for a full 7-day week (21 meals plus soft drinks, candy bars, or other in-between snacks) obtained by duplicating the meals of a different individual each day. Each institution supplies one complete 7-day, 21-meal diet sample each month. Each day's sample is kept frozen during the collection period. After collection, the total sample is packed in dry ice and shipped by air express to either the Southwestern Radiological Health Laboratory, Las Vegas, Nevada, the Southeastern Radiological Health Laboratory, Montgomery, Alabama, or the Northeastern Radiological Health Laboratory, Winchester, Massachusetts.

Each sample is packaged in three parts: (1) solid and semisolid food minus those portions not ordinarily eaten; (2) liquid milk; (3) other beverages such as soft drinks, coffee and tea. A record of the contents of each meal and the approximate weight of each item is made by the institution's dietician and sent with the sample. Samples usually range from 6 to 16 liters in volume and weigh from 8 to 20 kilograms.

### *Analytical Procedures*

Because calcium and phosphorous compounds may have an effect on the uptake of important bone-seeking radionuclides such as strontium-89 and strontium-90 (2), they are included in the analytical program. Total weight, stable calcium, and stable potassium determinations are obtained by conventional gravimetric or spectrophotometric methods. Phosphate determinations are made by a colorimetric technique.

The radioanalysis program is designed around three basic procedures: (1) gamma spectroscopy, (2) chemical separation of strontium-89 and strontium-90 with subsequent counting, and (3) total radium analysis. In the absence of interferences other than that from naturally-occurring radioactive potassium ( $K^{40}$ ), minimum detectable concentrations for the gamma scan, expressed as pc/kg are: iodine-131, 10; cesium-137, 5; and barium-140,

10 pc/kg. Approximate minimum detectable concentrations for strontium-89, strontium-90, and total radium are: 5, 1, and 1 pc/kg, respectively. Since a constant weight of food is analyzed, the minimum detectable level on a per day basis (pc/day) will be dependent on the food intake.

Total radium is determined by ashing, separation, and coprecipitation of radium as sulfate or chromate. After samples are transferred to planchets and dried, alpha activity is measured by an internal proportional counter with an appropriate delay for checking ingrowth of radium daughters. Since naturally-occurring radionuclides may contribute to the reported total radium values, the total radium technique is a practical screening indicator only. The bone dose, calculated by assuming total radium to be only radium-226, would therefore be moderately high.

### *Data*

Table 1 presents the dietary intake data expressed on a per-day basis from July 1963 through September 1963 for the 21 institutions from which samples were received. Also contained in the table is the range of ages of the children from which samples are being obtained. The reported radionuclide concentrations of these samples are extrapolated to the end of the sample collection period. The true iodine-131 intakes, therefore, may be somewhat greater than the reported values.

Certain of the radioanalyses are reported by the laboratories as being "less than" ( $<$ ) a specified value. For data averaging, the method employed for presentation in table 1 is that all "less-than" data are assumed to be equal to the full "less-than" values as they appear in the column entitled "monthly maximum averages." The column entitled "monthly minimum averages" reflects the averages in which all "less-than" values are considered to be zero.

Figure 2 shows the overall average daily intake of radionuclides at all institutions since January 1961. The data are also presented graphically in figure 3, as a distribution of all sample values observed during the three months versus daily intake. The number of values reported during this quarter in each range is plotted as a frequency-distribution step chart.

TABLE 1.—INSTITUTIONAL DAILY DIETARY INTAKE (BASED ON A 7-DAY COMPOSITE SAMPLE)

	Month (1963)	Alaska Palmer	California Los Angeles	Colorado Denver	Florida Tampa	Georgia Atlanta	Hawaii Honolulu	Illinois Chicago	Louisiana New Orleans	Massachusetts Boston	Minnesota Minneapolis	Missouri St. Louis
Age (years)		6-18	11-18	4-17	6-18	6-18	5-16	6-15	7-18	<sup>a</sup>	<sup>b</sup> <1-16	7-16
Total weight (kg/day)	July Aug Sept	1.21 1.76 1.33	— 1.20 —	2.48 1.89 2.47	1.94 1.91 2.91	1.43 1.63 1.81	1.87 1.70 1.26	1.79 1.53 1.88	2.12 3.13 2.42	1.45 1.48 1.66	1.56 1.83 1.56	— 2.74 2.51
Calcium (g/day)	July Aug Sept	0.5 0.9 0.6	— 1.2 —	1.5 1.2 1.5	1.1 1.1 1.1	0.5 0.6 0.6	0.6 0.9 0.6	1.2 1.1 1.3	1.3 1.3 1.0	0.9 1.0 1.2	0.7 0.7 0.6	— 0.6 1.6
Phosphorus as phosphate (g/day)	July Aug Sept	1.8 3.1 2.5	— 3.3 —	5.3 3.3 5.2	3.4 4.9 5.5	2.5 3.7 4.3	1.5 4.2 2.5	6.7 3.7 2.3	5.5 8.0 5.5	3.4 3.3 1.9	1.3 3.1 2.8	— 2.5 6.2
Potassium (g/day)	July Aug Sept	1.8 2.8 2.5	— 1.6 —	4.2 1.9 4.3	2.0 2.9 3.0	1.4 2.0 2.5	2.1 2.6 1.9	2.5 2.7 1.9	3.0 5.0 3.6	2.0 2.2 2.7	2.2 3.1 2.4	— 4.7 3.7
Total radium (pc/day)	July Aug Sept	<1 3.0 <1	— 2.0 —	3.0 1.0 <1	4.4 6.9 <3	2.7 <3.9 <4.0	<1 <1 <1	<1 <1 <1	<3 4.2 <4.4	<1 <1 1	<1 <1 <1	— 1.0 4.0
Strontium-89 (pc/day)	July Aug Sept	<5 30 10	— 10 —	<5 40 40	40 40 45	40 40 50	<5 5 5	50 15 25	130 195 185	5 30 45	15 45 35	— 5 40
Strontium-90 (pc/day)	July Aug Sept	14 25 17	— 12 —	79 33 16	15 36 32	8 31 25	17 13 7	100 22 26	34 64 48	18 45 43	26 18 18	— 11 57
Cesium-137 (pc/day)	July Aug Sept	90 265 105	— 40 —	210 65 135	280 345 470	95 165 165	85 85 65	145 120 185	180 360 230	225 320 315	125 185 135	— 150 150
Barium-140 (pc/day)	July Aug Sept	<10 <10 <10	— <10 —	<10 <10 <10	<20 <30 <30	<20 <30 <30	<10 <10 <10	<10 <10 <20	<30 <40 <30	<10 <10 <20	<10 <10 <10	— <10 <10
Iodine-131 (pc/day)	July Aug Sept	<10 <10 <10	— <10 —	<10 <10 <10	<20 <30 <30	<20 <30 <30	<10 <10 <10	<10 <10 <20	<30 <40 <30	<10 <10 <20	<10 <10 <10	— <10 <10

<sup>a</sup> Ages not available.<sup>b</sup> Food samples not collected from children too young for solid diet.

The number of stations used in constructing these graphs was 18, 19, and 20 for the months of July, August, and September 1963, respectively. Therefore, the total number of samples represented in each chart is 57.

#### Discussion of Data

Total intake ranged between 0.90 and 3.13 kg/day during this quarter. The frequency-distribution step chart shows that 74 percent of the samples weighed between one and two kg/day.

The calcium intake ranged between 0.5 and 1.7 g/day, with fifty-six percent of the values being one g/day or less. Phosphate intake ranged from 1.3 to 8.0 g/day, with 84 percent of the values being less than 5 g/day. Ninety-three percent of the samples analyzed for potassium showed that the intake was between 1 and 4 g/day.

Total radium intake ranged between <1 and 6.9 pc/day with all but two samples having less than 5 pc/day.

Strontium-89 intake ranged between <5 and 195 pc/day. Twenty-one percent of the samples gave values between <5 and 20 pc/day. For purposes of comparison the Federal Radiation Council (FRC) range II for strontium-89 is 200 to 2000 pc/day (3). The maximum strontium-90 intake during this quarter was 100 pc/day. Again for comparison, 33 percent of the values were below 20 pc/day. The FRC range II for strontium-90 is 20 to 200 pc/day (3, 4).

Although the intake of cesium-137 ranged from 40 to 470 pc/day, the distribution shows a pronounced peak between 120 and 160 pc/day formed by 32 percent of the results.

Both barium-140 and iodine-131 were below the limits of detectability during this quarter.

TABLE 1.—INSTITUTIONAL DAILY DIETARY INTAKE (BASED ON A 7-DAY COMPOSITE SAMPLE)  
Continued—

	Month (1963)	Montana Helena	Nebraska Omaha	New Mexico Albuquerque	New York New York	Ohio Cleveland	Tennessee Memphis	Texas Austin	Utah Salt Lake City	Virginia Norfolk	Washington Seattle	Monthly minimum average	Monthly maximum average
Age (years)		6-17	6-18	5-15	8-15	6-15	8-18	6-18	12-18	10-18	6-16		
Total weight (kg/day)	July Aug Sept	1.14 1.85 1.13	2.05 1.96 1.35	0.90 1.73 1.89	1.60 — 1.57	1.73 1.74 1.86	1.78 2.00 1.65	2.25 1.97 2.10	— — 1.87	1.99 1.78 1.72	2.44 2.02 1.79	1.76 1.89 1.84	1.76 1.87 1.84
Calcium (g/day)	July Aug Sept	0.5 0.8 0.9	1.2 1.2 0.7	1.3 1.6 1.1	0.6 — 0.6	1.2 1.0 1.2	1.2 0.9 0.9	1.7 0.8 0.8	— — 1.2	0.9 0.7 0.6	1.3 1.0 0.7	1.0 1.0 0.9	1.0 1.0 0.9
Phosphorus as phosphate (g/day)	July Aug Sept	2.1 3.6 2.9	1.7 3.9 2.8	1.5 4.0 4.2	4.4 — 3.2	4.0 7.5 3.1	4.0 4.6 4.3	7.0 4.9 4.9	— — 3.7	4.6 4.0 3.7	4.9 4.5 2.6	3.5 4.2 3.7	3.5 4.2 3.7
Potassium (g/day)	July Aug Sept	1.8 3.3 2.0	3.1 3.1 2.1	1.3 2.9 2.7	3.3 — 3.2	3.0 3.0 3.1	2.4 3.4 1.9	2.9 2.8 2.4	— — 2.7	2.2 2.4 1.8	3.9 3.4 3.0	2.5 2.9 2.7	2.5 2.9 2.7
Total radium (pc/day)	July Aug Sept	<1 3.0 <1	1.0 1.0 2.0	2.0 1.0 <1	<1 — 1.0	1.0 <1 1.0	<4 3.1 <2.2	<3 3.6 <5.1	— — <1	<3 2.5 <4.0	<1 1.0 <1	4.1 1.4 0.5	5.2 2.2 2.1
Strontium-89 (pc/day)	July Aug Sept	55 40 20	25 40 20	<5 15 5	30 — 5	20 40 15	125 70 45	70 35 30	— — 30	55 30 45	40 45 15	39 41 32	40 41 36
Strontium-90 (pc/day)	July Aug Sept	16 26 18	33 27 2	10 15 13	27 — 28	29 77 25	26 41 40	29 24 24	— — 23	25 22 20	32 33 30	30 30 26	30 30 26
Cesium-137 (pc/day)	July Aug Sept	150 190 175	125 145 80	25 50 75	120 — 135	145 170 135	135 125 85	130 105 65	— — 75	105 95 125	270 235 215	147 169 156	147 169 156
Barium-140 (pc/day)	July Aug Sept	<10 <10 <10	<10 <10 <10	<10 <10 <10	<10 — 10	<10 <10 <20	<30 <30 <20	<30 <30 <30	— — <10	<30 <30 <30	<10 <10 <10	0 0 0	16 17 16
Iodine-131 (pc/day)	July Aug Sept	<10 <10 <10	<10 <10 <10	<10 <10 <10	<10 — <10	<10 <10 <20	<30 <30 <20	<30 <30 <30	— — <10	<30 <30 <30	<10 <10 <10	0 0 0	16 17 16

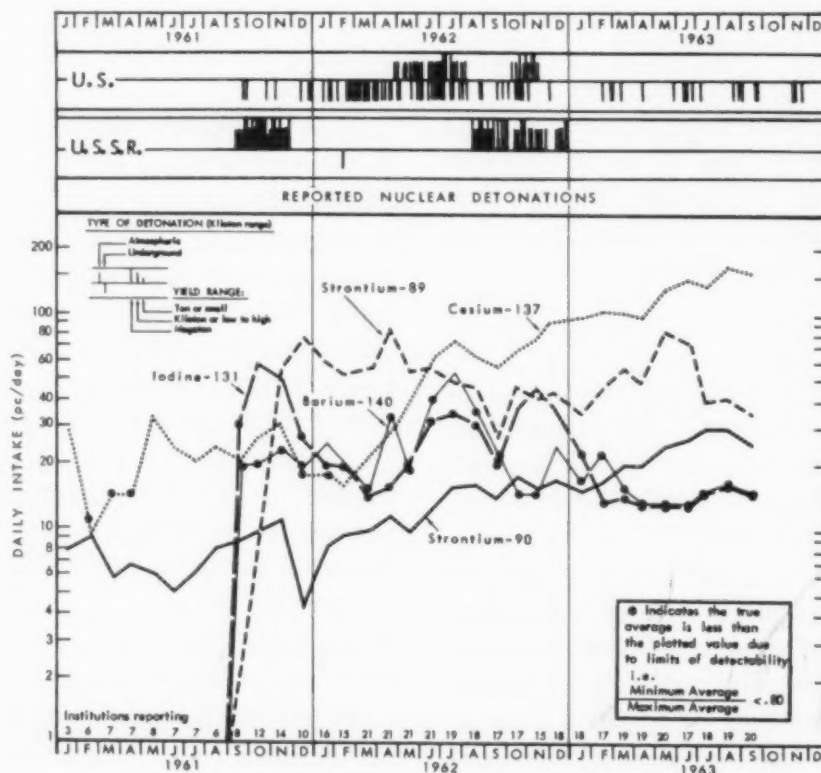


FIGURE 2.—RADIONUCLIDES IN INSTITUTIONAL DIET SAMPLES—AVERAGE OF INSTITUTIONS

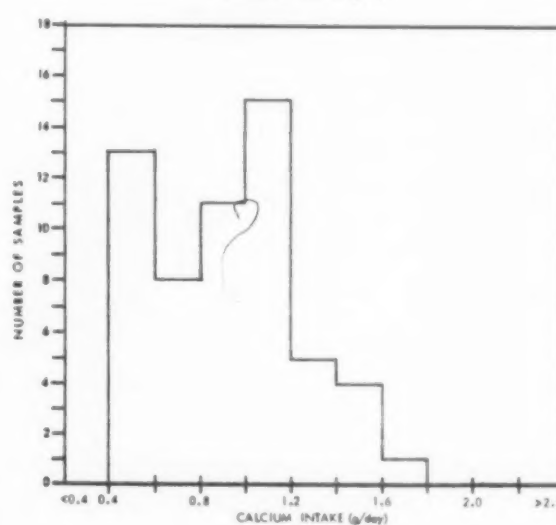
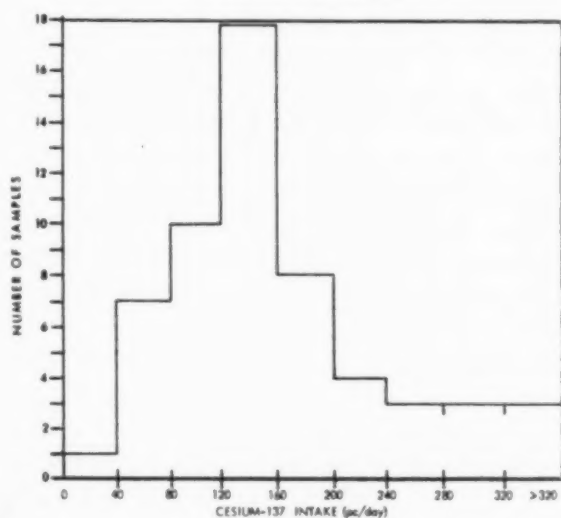
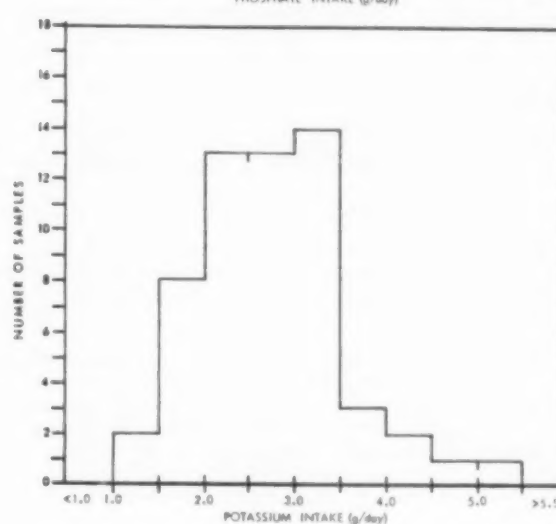
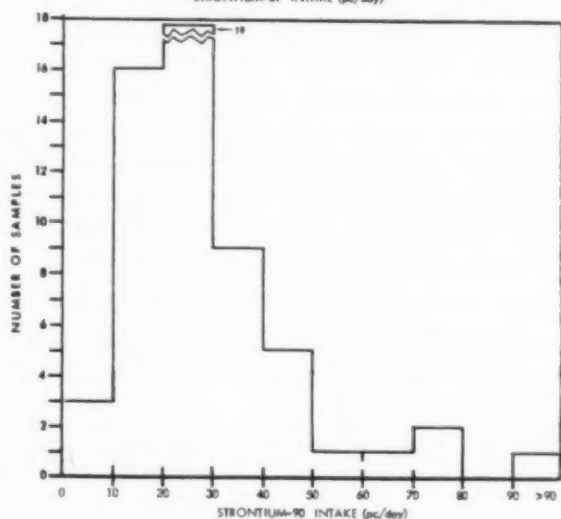
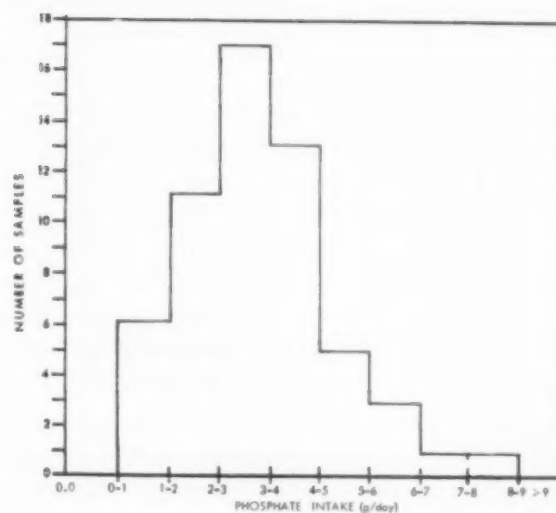
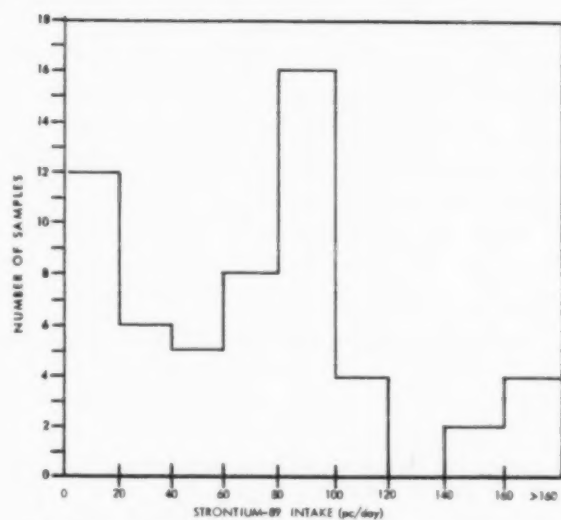


FIGURE 3.—DISTRIBUTION OF THE NUMBER OF SAMPLES VERSUS INTAKE OF MATERIALS IN INSTITUTIONAL DIETS FOR JULY—SEPTEMBER 1963

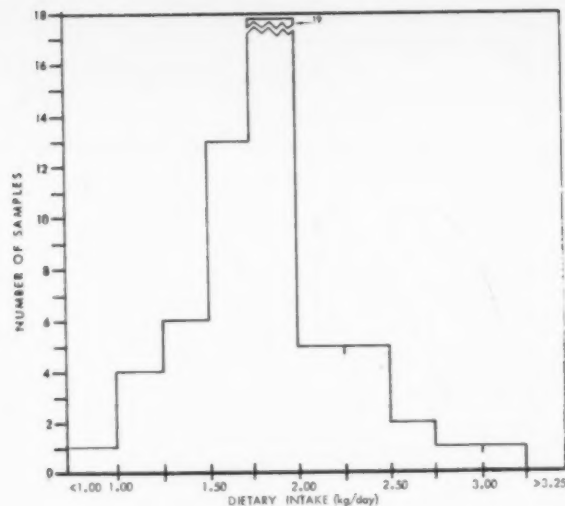
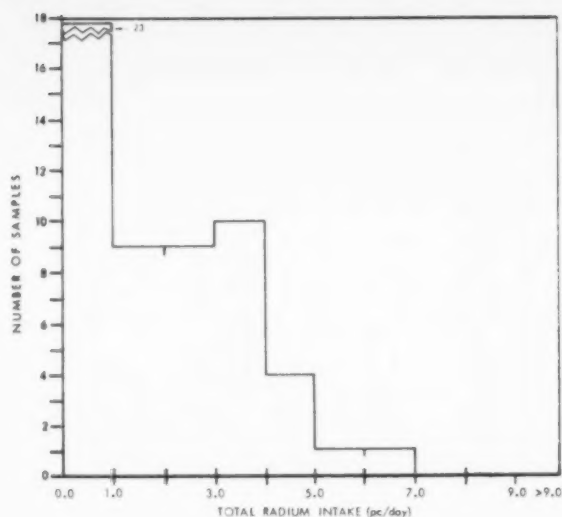


FIGURE 3.—DISTRIBUTION OF THE NUMBER OF SAMPLES VERSUS INTAKE OF MATERIALS IN INSTITUTIONAL DIETS FOR JULY—SEPTEMBER 1963—  
Continued

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#### Previous coverage in *Radiological Health Data*:

Period	Issue
March-June 1962	December 1962
July-September 1962	April 1963
October-December 1963	July 1963
January-March 1963	September 1963
April-June 1963	December 1963

# TRI-CITY DIET STUDY,<sup>1</sup> MAY-JULY 1963

Health and Safety Laboratory, AEC

Since March 1960, the Health and Safety Laboratory, through its quarterly diet study, has made estimates of the strontium-90 content of the average diet of individuals living in New York City, San Francisco, and Chicago.

Selected foods representing 19 food categories are purchased at each of these three cities about every 3 months and are analyzed for strontium-90. Using data from the U. S. Department of Agriculture (1), the annual consumption by an average individual can be grouped into the same 19 food categories. The annual dietary intake of strontium-90 can be estimated by summing the contributions from each category. Some food types are assumed to be representative of larger food categories, such as liquid milk for dairy products in general.

The consumption data from the "Household Food Survey of 1955" are based on a weight-as-purchased basis. Before the food samples for the Tri-City Diet Study are ashed for radiochemical analysis, they are prepared to a certain degree as if for actual consumption. For example, fruits are peeled, eggs are shelled, and poultry is boned. Therefore, concentrations of radioactivity in foods as reported in the Tri-City Diet Study are based on the trimmed weight. No correction is made for the waste.

After two samplings at each city it was found that the calcium content of most food categories did not vary among cities, nor did it vary significantly with time. Calcium analyses were therefore discontinued and average calcium content of foods was computed and used to estimate the average annual intake of this mineral. The specific numbers used to calculate calcium intake are given in HASL-113 (2).

Results obtained from the programs thirteenth sampling May-July 1963 are presented in table 1. The variation with time of the daily intake of strontium-90 in the three cities is plotted in figure 1.

<sup>1</sup> Summarized from *Fallout Program Quarterly Summary Report, HASL 142:182-4*, Office of Technical Services, Department of Commerce, Washington 25, D. C., (January 1964), price \$4.00.

## Discussion

The previously noted geographic pattern of distribution of strontium-90 in the diet is seen to persist in the last sampling. Levels have been highest in New York City and lowest in San Francisco. Partly due to its high annual consumption, milk continues to be the predominant source of strontium-90 in the diet.

The sharp increases in the daily intake of strontium-90 at each of the cities between the first and second quarters of 1963 are evident in figure 1. These increases were due principally to the higher strontium-90 content of milk. Dairy cows in the milksheds of the three cities were put out to pasture during this period and presumably consumed grass heavily contaminated with strontium-90 deposited during the spring and early summer.

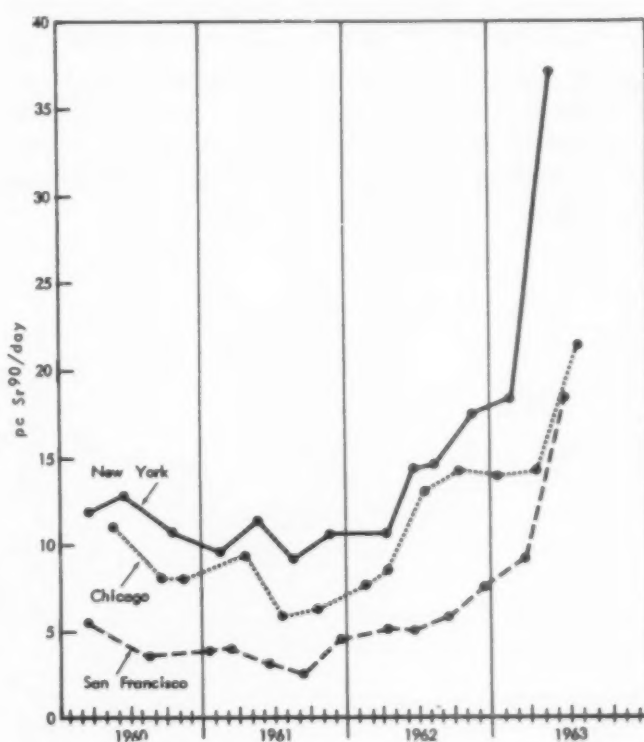


FIGURE 1.—DAILY INTAKE OF STRONTIUM-90 IN TRI-CITY TOTAL DIETS

## REFERENCES

- (1) U. S. Department of Agriculture: *Food Consumption of Households in the United States, Report No. 1*

TABLE 1.—AVERAGE PER PERSON DIETARY CONSUMPTION AND STRONTIUM-90 INTAKE—  
THIRTEENTH SAMPLING

Food category	Average U. S. consumption		New York City May 1963		Chicago July 1963		San Francisco June 1963	
	diet (kg/yr)	Calcium (g/yr)	pc/kg <sup>a</sup>	pc/yr	pc/kg	pc/yr	pc/kg	pc/yr
Bakery products.....	37	37.0	15.0±2.8	555	18.4±1.0	679	12.0±1.1	445
Whole grain products.....	11	10.0	51.1±1.8	562	47.2±2.0	519	18.6±1.6	204
Eggs.....	16	9.1	3.4±0.1	54	9.4±0.2	150	5.9±0.2	95
Fresh vegetables.....	43	15.0	20.3±0.6	873	6.4±0.6	276	11.2±0.6	482
Root vegetables.....	17	6.1	7.6±0.5	129	4.1±0.4	70	4.0±0.5	67
Milk.....	221	234.3	40.8±1.0	9017	17.1±0.7	3777	19.3±0.7	4263
Poultry.....	17	9.2	0.7±0.1	12	2.9±0.2	50	3.5±0.2	60
Fresh fish.....	8	10.8	0.8±0.1	6	1.3±0.1	10	0.8±0.1	6
Flour.....	43	8.6	23.5±0.6	1011	28.3±0.7	1215	7.0±0.3	301
Macaroni.....	3	0.7	14.0±0.6	42	16.8±0.6	50	12.3±0.7	37
Rice.....	3	1.1	1.8±0.3	5	4.1±0.4	12	2.8±0.3	8
Meat.....	73	10.9	0.7±0.2	51	2.3±0.1	171	2.4±0.2	176
Shellfish.....	1	0.8	2.8±0.5	3	1.2±0.1	1	1.5±0.3	2
Dried beans.....	3	2.9	14.0±1.8	42	lost	90	6.5±1.9	20
Fresh fruit.....	68	13.6	11.1±3.8	755	2.8±0.3	190	3.0±0.3	203
Potatoes.....	45	5.8	5.8±0.6	216	2.3±0.2	104	1.1±0.4	49
Canned fruit.....	26	1.3	1.4±0.2	36	1.6±0.2	42	1.2±0.1	30
Fruit juices.....	19	1.7	2.3±0.3	44	3.3±0.3	62	3.8±0.3	72
Canned vegetables.....	20	4.2	6.5±0.5	130	8.1±0.6	162	1.9±0.4	37
Annual Intake.....	674	383		13,543		7,630		6,557
pc Sr <sup>90</sup> /g Ca in total diet.....				35.3		20.0		17.1

<sup>a</sup> Error terms are one standard deviation (due to counting).

<sup>b</sup> Estimated from April 1963 sampling.

(1955), *Household Food Consumption Survey*, Superintendent of Documents, Government Printing Office, Washington 25, D. C. (December 1956), price \$1.00.

(2) U. S. Atomic Energy Commission: *Fallout Program Quarterly Summary Report*, HASL-113:85-89, Office of Technical Services, Department of Commerce, Washington 25, D. C. (July 1, 1961), price \$2.50.

#### Recent coverage in *Radiological Health Data*:

Period	Issue
Eighth sampling (April 1962)	January 1963
Ninth sampling (June-July 1962)	March 1963
Tenth sampling (August-September 1962)	June 1963
Eleventh sampling (November 1962-January 1963)	September 1963
Twelfth sampling (February-April 1963)	December 1963

## Section III.—Water

### RADIOACTIVITY IN SURFACE WATERS OF THE UNITED STATES, SEPTEMBER 1963

*Division of Water Supply and Pollution Control, Public Health Service*

Levels of radioactivity in surface waters of the United States have been under surveillance by the Public Health Service National Water Quality Network since its initiation in 1957. Beginning with the establishment of 50 sampling points, this network has expanded to 128 stations as of February 1, 1964. These are operated jointly with other Federal, State, and local agencies, and industry. Samples are taken from surface waters of all major U. S. river basins for physical, chemical, biological and

radiological analyses. These data can be used for evaluating sources of radioactivity which may affect specific domestic, commercial, and recreational uses of surface water. Further, the Network provides background information necessary for recognizing pollution and water quality trends and for determining levels of radioactivity to which the population may be subjected. Data assembled through the Network are published in an annual compilation (1-6).



FIGURE 1.—TOTAL BETA ACTIVITY (pc/liter) IN SURFACE WATER AT NATIONAL WATER QUALITY NETWORK SAMPLING STATIONS, SEPTEMBER 1963

## Sampling Procedures

The participating agencies collect one-liter "grab" samples each week and ship them to the Public Health Service laboratory in Cincinnati for analysis. Determinations for gross alpha and gross beta radioactivity in the suspended and dissolved solids and of strontium-90 activity in the total solids are carried out on frequency schedules based on need.

Gross beta activity in each weekly sample was determined until January 1960, when the levels became essentially equal to background. Thereafter, gross beta determinations were made on monthly composites of the weekly samples received from all stations, except those located downstream from known potential sources of radioactive waste and those from all newly established Network stations. (Weekly alpha and beta measurements are scheduled routinely during the first year of operation at newly established stations.) On September 1, 1961, weekly determinations of gross beta activity again were instituted to permit rapid detection of activity due to fallout from renewed weapons testing. This practice was continued until the end of October 1962, when samples for gross beta analysis were again composited monthly. Gross alpha determinations were made once a month except where variable or high values observed during the first year indicated the need for more frequent measurement.

Normally, samples are counted at the Cincinnati Laboratory within two weeks following collection or within one week after compositing. The decay of activity is followed on each sample when the first analysis shows unusually high activity. Also, if a recount indicates that the original analysis was questionable, values based on recounting are recorded. All results are reported for the time of counting and are not corrected by extrapolation to the time of collection.

## Analytical Methods

The analytical method used for determining gross alpha and beta radioactivity is described in the eleventh edition of "Standard Methods for the Examination of Water and Wastewater" (7). Suspended and dissolved solids are separated by passing the sample through a membrane filter (type HA) with a pore size of 0.45 microns. Planchets are then prepared for count-

ing the dissolved solids (in the filtrate) and the suspended solids (on the charred membrane filter) in an internal proportional counter. Reference sources of  $U_3O_8$ , which give a known count rate if the instrument is in proper calibration, are used for daily checking of the counters.

Since the fourth quarter of 1958, strontium-90 analyses have been made on three-month composites of aliquots from weekly samples. Beginning November 1962 the frequency of strontium-90 analyses was reduced to twice a year at each sampling point except those stations immediately below nuclear installations. Until the fourth quarter of 1961, the method used for determining strontium-90 was that described in the above reference (7). Tributyl phosphate was used to extract ingrown yttrium-90 from the purified, coprecipitated strontium-90. Beginning with the first quarter of 1962, a modification of a procedure described by Harley has been used (8). The yttrium-90, together with an yttrium carrier, is precipitated at pH 8.5; the precipitate is washed, redissolved, and reprecipitated as yttrium oxalate and the latter is washed and counted in a low-background, anti-coincidence, end-window proportional counter.

## Results

Table 1 presents September 1963 results of alpha and beta analyses of U. S. raw surface waters. The stations on a river are arranged in the table according to their relative location on the river, the first station listed being closest to the headwaters. These data are preliminary; reanalysis of some samples and some analyses which are not completed at the time of this report will be included in the Network's Annual Compilation of Data (6). The figures for gross alpha and gross beta radioactivity represent either determinations made on composite samples or means of weekly determinations where composites were not made. The monthly means are reported to the nearest pc/liter with the exception that when all samples have zero pc/liter the mean is reported as zero. When the calculated mean is between zero and 0.5 the mean is reported as  $< 1$  pc/liter. The most recent quarterly strontium-90 results appeared in the January 1964 *Radiological Health Data* (9).

TABLE 1.—RADIOACTIVITY IN U.S. SURFACE WATERS, SEPTEMBER 1963

[Average concentrations in pc/liter]

Station	Beta activity			Alpha activity			Station	Beta activity			Alpha activity		
	Sus-pended	Dis-solved	Total	Sus-pended	Dis-solved	Total		Sus-pended	Dis-solved	Total	Sus-pended	Dis-solved	Total
Allegheny River:							Monongahela River:						
Pittsburgh, Pa.	9	21	30	0	0	0	Pittsburgh, Pa.	0	20	20	0	0	0
Animas River:							North Platte River:						
Cedar Hill, N. Mex.	6	10	16	1	1	2	Henry, Nebr.	10	37	47	1	22	23
Apalachicola River:							Ohio River:						
Chattahoochee, Fla.	1	16	17	0	0	0	Toronto, Ohio	2	23	25	0	1	1
Arkansas River:							Addison, Ohio	5	32	37	0	0	0
Coolidge, Kansas	272	43	315	24	21	45	Huntington, W. Va.	2	14	16	1	1	2
Ponca City, Okla.	98	64	162	10	2	12	Cincinnati, Ohio	0	20	20	0	1	1
Fort Smith, Ark.	266	34	300	46	0	46	Louisville, Ky.	4	29	33	0	0	0
Little Rock, Ark.	29	38	67	3	1	4	Evansville, Ind.	2	21	23	1	1	2
Pendleton Ferry, Ark.	3	15	18	1	2	3	Cairo, Ill.	0	19	19	0	<1	<1
Bear River:							Ouachita River:						
Preston, Idaho	5	44	49	0	1	1	Bastrop, La.	7	23	30	2	1	3
Big Horn River:							Pend Oreille River:						
Hardin, Mont.	405	51	456	61	9	70	Albeni Falls Dam,						
Big Sioux River:							Idaho	1	11	12	0	0	0
Sioux Falls, S. Dak.	34	24	58	1	0	1	Platte River:						
Chattahoochee River:							Plattsmouth, Nebr.	124	46	170	10	2	12
Atlanta, Ga.	8	11	19	0	0	0	Potomac River:						
Columbus, Ga.	0	7	7	0	0	0	Williamsport, Md.	0	12	12	0	2	2
Lanett, Ala.	12	11	23	1	0	1	Great Falls, Md.	0	19	19	0	0	0
Chena Slough:							Washington, D. C.	2	27	29	0	1	1
Fairbanks, Alaska	3	6	9	0	0	0	Rainy River:						
Clearwater River:							Baudette, Minn.	7	47	54	0	0	0
Lewiston, Idaho	4	9	13	1	0	1	International Falls,						
Clinch River:							Minnesota	8	49	57	0	0	0
Clinton, Tenn.	2	6	8	0	<1	<1	Raritan River:						
Kingston, Tenn.	6	42	48	0	0	0	Perth Amboy, N. J.						
Colorado River:							(5-ft. Below Surface)	4	13	17	0	3	3
Loma, Colo.	175	44	219	16	7	23	Perth Amboy, N. J.						
Page, Ariz.	3	42	45	1	7	8	(5-ft. Above Bottom)	2	10	12	0	3	3
Boulder City, Nev.	0	13	13	0	8	8	Red River, North:						
Parker Dam, Calif-Ariz.	4	13	17	0	6	6	Grand Forks, N. Dak.	13	37	50	0	0	0
Yuma, Ariz.	0	288	288	0	0	0	Red River, South:						
Columbia River:							Denison, Tex.	0	23	23	0	0	0
Northport, Wash.	4	21	25	1	1	2	Index, Ark.	4	35	39	0	1	1
Wenatchee, Wash.	0	14	14	0	1	1	Bossier City, La.	5	35	40	0	0	0
Pasco, Wash.	42	662	704	0	0	0	Alexandria, La.	4	26	30	0	3	3
McNary Dam, Ore.	34	332	366	0	1	1	Rio Grande River:						
Bonneville, Ore.	16	293	309	0	1	1	Alamosa, Colo.	26	77	103	1	9	10
Clatskanie, Ore.	12	204	216	0	<1	<1	El Paso, Tex.	87	21	108	12	3	15
Cumbarland River:							Laredo, Tex.	694	45	739	47	3	50
Clarksville, Tenn.	6	15	21	1	0	1	Brownsville, Tex.	7	21	28	0	3	3
Connecticut River:							Roanoke River:						
Wilder, Vt.	15	18	33	0	0	0	John H. Kerr						
Northfield, Mass.	24	18	42	1	0	1	Reser/Dam, Va.	2	10	12	1	0	1
Enfield Dam, Conn.	9	15	24	0	0	0	Sabine River:						
Cuyahoga River:							Ruliff, Tex.	11	46	57	0	1	1
Cleveland, Ohio	4	31	35	0	2	2	Sacramento River:						
Delaware River:							Courtland, Calif.	98	15	113	2	0	2
Martins Creek, Pa.	2	9	11	0	0	0	San Joaquin River:						
Trenton, N. J.	1	16	17	0	0	0	Vernalis, Calif.	12	25	37	1	5	6
Philadelphia, Pa.	8	18	26	1	1	2	San Juan River:						
Escambia River:							Shiprock, N. Mex.	120	26	146	18	10	28
Century, Fla.	7	5	12	1	0	1	St. Lawrence River:						
Great Lakes:							Massena, N. Y.	4	13	17	1	1	2
Duluth, Minn.	3	8	11	0	0	0	Schuykill River:						
Sault Ste. Marie, Mich.	1	5	6	0	1	1	Philadelphia, Pa.	3	24	27	0	0	0
Milwaukee, Wis.	3	15	18	0	1	1	Savannah River:						
Gary, Ind.	2	11	13	0	0	0	North Augusta, S. C.	12	16	28	0	0	0
Port Huron, Mich.	4	9	13	0	0	0	Port Wentworth, Ga.	4	16	20	<1	0	<1
Detroit, Mich.	4	11	15	0	0	0	Shenandoah River:						
Buffalo, New York	9	21	30	0	0	0	Berryville, Va.	15	14	29	0	0	0
Green River:							Ship Creek:						
Dutch John, Utah	4	37	41	0	3	3	Anchorage, Alaska	2	10	12	0	0	0
Hudson River:							Snake River:						
Poughkeepsie, N. Y.	4	28	32	0	0	0	Ice Harbor Dam,						
Illinois River:							Washington	3	19	22	0	0	0
Peoria, Ill.	3	37	40	0	0	0	Wawawai, Wash.	6	20	26	0	3	3
Grafton, Ill.	198	24	162	21	0	21	Payette, Idaho	7	15	22	0	4	4
Kanawha River:							South Platte River:						
Winfield Dam, W. Va.	8	22	30	0	0	0	Julesburg, Colo.	18	58	76	4	32	36
Klamath River:							Spokane River:						
Keno, Ore.	7	17	24	0	1	1	Post Falls, Idaho	3	9	12	0	0	0
Kansas River:							Susquehanna River:						
De Soto, Kans.	78	72	150	7	5	12	Sayre, Pa.	6	12	18	0	0	0
Maumee River:							Conowingo, Md.	0	17	17	0	1	1
Toledo, Ohio	6	27	33	1	1	2	Tennessee River:						
Little Miami River:							Lenoir City, Tenn.	3	17	20	0	0	0
Cincinnati, Ohio	16	13	29	2	2	4	Chattanooga, Tenn.	1	21	22	1	0	1
Merrimack River:							Bridgeport, Ala.	0	17	17	0	0	0
Lowell, Mass.	12	26	38	0	0	0	Pickwick Landing,						
Mississippi River:							Tenn.	5	24	29	1	0	1
St. Paul, Minn.	8	43	51	0	2	2	Tombigbee River:						
Dubuque, Iowa	4	35	39	0	1	1	Columbus, Miss.	14	34	48	1	1	2
Burlington, Iowa	7	27	34	0	1	1	Truckee River:						
E. St. Louis, Ill.	8	34	42	0	2	2	Farad, Calif.	4	14	18	0	0	0
Cape Girardeau, Mo.	14	44	58	1	3	4	Wabash River:						
W. Memphis, Ark.	4	27	31	0	2	2	New Harmony, Ind.	4	24	28	0	5	5
Vicksburg, Miss.	15	24	39	1	1	2	Willamette River:						
Delta, La.	16	32	48	0	2	2	Portland, Ore.	6	3	9	0	0	0
New Orleans, La.	1	25	26	0	2	2	Yakima River:						
Missouri River:							Richland, Wash.	0	13	13	0	2	2
Williston, N. Dak.	11	39	50	1	5	6	Yellowstone River:						
Bismarek, N. Dak.	7	23	30	0	4	4	Sidney, Mont.	239	45	284	7	4	11
Yankton, S. Dak.	0	33	33	0	5	5							
Omaha, Nebr.	27	30	57	9	4	13	Maximum	694	662	739	61	32	70
St. Joseph, Mo.	50	32	82	8	4	12	Minimum	0	3	6	0	0	0
Kansas City, Kans.	31	35	66	9	4	13							
Missouri City, Mo.	27	45	72	3	5	8							
St. Louis, Mo.	14	35	49	2	0	2							

Note: These data are preliminary; reanalysis of some samples and analyses which are not complete at the time of this report will be included in the network's Annual Compilation of Data (6).

In order to obtain a geographical perspective of the radioactivity in surface water, the numbers alongside the various stations in figure 1 give the September 1963 average total beta activity in suspended-plus-dissolved solids in raw water collected at that station. Network results for the years 1957-1962 have been summarized by Weaver *et al* (10).

### Discussion

Analysis of the preliminary September 1963 data indicates several elevated levels of alpha and beta activity (pc/liter) in the suspended solids fraction. The monthly average suspended alpha and beta activities in the Rio Grande River at Laredo, Texas were 47 and 694 pc/liter, respectively. Samples from other stations on this river indicate no unusual values. In the Big Horn River at Hardin, Montana, the monthly average was 61 pc/liter of suspended alpha activity and 405 pc/liter of suspended beta activity. The dissolved alpha and beta activities were not significantly different from those commonly observed at these sampling points. The samples associated with these unusually high averages contained large quantities of suspended solids. Thus, only quite small aliquots could be used in the laboratory determinations to avoid excessive self-absorption. In such cases the relatively large multiplication factor, together with the usual range of counting errors associated with standard counting periods, can result in apparently abnormal levels of radioactivity. The specific activity (pc/g) of the samples discussed above was determined and found to be comparable to the specific activity observed normally in samples from these stations. The apparent increased activity, therefore, may be attributed to the extremely high suspended solids content of the samples and essentially of natural origin.

On the Columbia and Colorado Rivers, the monthly dissolved beta activity averages were greater than 100 pc/liter. Of the six stations on the Columbia River, the four downstream from the Hanford Atomic Products Operation facility had averages of between 204 and 662 pc/liter. It was observed that the concentration diminished progressively downstream from the facility. An average of 288 pc/liter of dissolved beta activity was recorded on the Colorado River at Yuma, Arizona. Other stations

on this river reported concentrations not nearly as high. While there are no generally applicable standards for surface water, the radioactivity associated with dissolved solids provides a rough indication of the levels which could occur in treated water, since nearly all suspended matter is removed by the treatment process (11). The Public Health Service Drinking Water Standards state that in the absence of strontium-90 and alpha emitters,<sup>1</sup> a water supply is acceptable when the gross beta concentrations do not exceed 1000 pc/liter (12).

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- (12) Public Health Service: *Drinking Water Standards, Revised 1962*, Public Health Service Publication No. 956, Superintendent of Documents, Government Printing Office, Washington, D. C. 20402. (March 1963), price 30 cents.

<sup>1</sup> Absence is taken here to mean a negligibly small fraction of the specific limits of 3 pc/liter and 10 pc/liter for unidentified alpha emitters and strontium-90, respectively.

<sup>2</sup> Single free copies of this publication may be obtained from: Public Inquiries Branch, Public Health Service, U. S. Department of Health, Education and Welfare, Washington, D. C. 20201.

# RADIOACTIVITY IN CALIFORNIA SURFACE WATER,<sup>1</sup> JANUARY-JUNE 1963

*Bureau of Radiological Health, State of California Department of Public Health*

Results obtained by the Bureau of Radiological Health in its monitoring of California surface water during the period January to June 1963 are summarized below. The importance of this facet of the Bureau's environmental surveillance program stems from the fact that most of California's domestic water supplies come from surface sources. Radioactivity in such water supplies consists of the natural radioactivity in surface streams and any radioactivity that may be added by the discharge of sewage or industrial waste effluents into streams. These water supplies may also contain radioactivity from fallout, particularly fallout into open terminal or distribution reservoirs. Most of the supplies sampled represent raw surface waters, although a few wells, along with some water supplies that use infiltration galleries, are also sampled (figure 1).

It is necessary to monitor domestic water supplies on a continuing basis, since it is impossible to forecast levels of radioactivity in these supplies on the basis of radioactivity in rain, snow, or surface streams. The Bureau has established a monthly sampling schedule whereby 500-ml samples are collected and the total solids are analyzed for alpha and beta radioactivity. In addition, a three-liter sample is collected each month for a period of six months to make up a composite of approximately five gallons for strontium-90 analysis.

## *Laboratory Methods.*

Radionuclide analyses of water are carried out in the Sanitation and Radiation Laboratory. All measurements of alpha and alpha-plus-beta activities are made with windowless gas-flow proportional counters. Five proportional counters, four of which are manual counters, are available to the Bureau. Two of the manual units have specially-designed, shielded external detectors which reduce the alpha and beta backgrounds to 0.01 and 30

cpm, respectively. In the case of the two integrally-constructed manual units the relatively high beta background has been reduced to 40 cpm by partial shielding of the scaler with lead bricks. The fifth counter is an automatic unit which has a beta background of about 15 cpm.

The Department's maximum capacity for alpha and beta radioactivity measurements during a normal work week on these instruments is 500 thirty-minute counts. Counting methods used are in accordance with U. S. Public Health Service recommended procedures (1).

## *Discussion*

Table 1 shows the monthly average beta activity in the suspended-plus-the-dissolved-solids in raw surface water in California from Janu-



FIGURE 2.—CALIFORNIA SURFACE WATER SAMPLING STATIONS

<sup>1</sup> "Radiological Health News," Vol 2 Nos. 3 and 4, July and October 1963, State of California, Department of Public Health, Bureau of Radiological Health, 2151 Berkeley Way, Berkeley 4, California.

TABLE 1.—GROSS BETA ACTIVITY IN CALIFORNIA SURFACE WATER, JANUARY-JUNE 1963

[Concentrations in pc/liter]

Sampling station	Jan.	Feb.	Mar.	Apr.	May	June
Antioch.....	27.7	99.4	59.9	97.0	35.7	73.9
Chula Vista.....	76.6	a				
Clearlake Highlands.....	0 <sup>b</sup>	80.0	0 <sup>b</sup>	24.8 <sup>b</sup>		87.8
Crescent City.....	0 <sup>b</sup>	225	0 <sup>b</sup>	0 <sup>b</sup>	7.5 <sup>b</sup>	43.6
Escondido.....	31.7	53.2	32.6	24.2 <sup>b</sup>	29.5	28.9
Eureka.....	25.9	0 <sup>b</sup>	0 <sup>b</sup>	46.5	8 <sup>b</sup>	
Fort Bragg.....	0 <sup>b</sup>	0 <sup>b</sup>	0 <sup>b</sup>	39.9	13.7 <sup>b</sup>	2.1 <sup>b</sup>
Fresno.....						
Lake Millerton.....	0 <sup>b</sup>	44.6 <sup>b</sup>	73.9 <sup>b</sup>	29.5	31.5	69.6
Marin Municipal Water District.....						
Nicasio Reservoir.....	0 <sup>b</sup>	106	91.9			
Mariposa.....	24.3	92.5	0 <sup>b</sup>	108.2	88.1	35.8
Metropolitan Water Company of Southern California.....						
Lake Havasu.....	0 <sup>b</sup>	0 <sup>b</sup>	25.5		0 <sup>b</sup>	18.5
Lake Matthews.....	0 <sup>b</sup>	0 <sup>b</sup>		11.6 <sup>b</sup>	30.0	11.6 <sup>b</sup>
Monterey.....	0 <sup>b</sup>	22.2	42.2	50.1	9.1 <sup>b</sup>	11.1 <sup>b</sup>
Napa.....	0 <sup>b</sup>				38.2	19.5 <sup>b</sup>
North Marin County Water District.....				56.0	68.5	47.0
Oroville.....						
Wyandotte Irrigation District.....		40.5		163.2		24.7 <sup>b</sup>
California Water Service.....	30.2		39.8		27.4	
Placerville.....					39.2	70.7
Redding.....	0 <sup>b</sup>	48.0	0 <sup>b</sup>	55.2	38.1	47.5
Sacramento.....	28.1	78.6	31.8	30.3	33.7	
San Francisco Water Department.....						
Alameda East.....	0 <sup>b</sup>	57.7	33.8	94.3	33.0	77.9
Brightside Weir.....	0 <sup>b</sup>	0 <sup>b</sup>	0 <sup>b</sup>	36.4	14.6 <sup>b</sup>	100.5
Calaveras Reservoir.....	0 <sup>b</sup>	81.4	0 <sup>b</sup>	18.1 <sup>b</sup>	50.1	24.7
Crystal Springs Raw Outlet.....	0 <sup>b</sup>	28.4	0 <sup>b</sup>	26.0		81.4
Crystal Springs Line 1.....	13.9 <sup>c</sup>	27.9 <sup>c</sup>	32.2 <sup>c</sup>	41.1 <sup>b</sup>	42.9 <sup>b</sup>	28.9 <sup>b</sup>
Crystal Springs Line 2.....					76.5	
Hetch Hetchy.....			61.1		186.6	71.6
Lombard Reservoir.....	0 <sup>b</sup>	93.3	23.5	21.0 <sup>b</sup>		
San Andreas Line 2.....	9.8 <sup>c</sup>	49.3 <sup>c</sup>	41.3 <sup>c</sup>	57.8 <sup>c</sup>	42.8 <sup>c</sup>	50.6 <sup>c</sup>
University Mound.....	0 <sup>b</sup>	0 <sup>b</sup>	22.6	70.9		
San Jose.....		0 <sup>b</sup>	30.3	0 <sup>b</sup>	38.9	
San Luis Obispo.....	0 <sup>b</sup>	118.3				
Santa Barbara.....	0 <sup>b</sup>	0 <sup>b</sup>	0 <sup>b</sup>	42.5	14.8 <sup>b</sup>	
Santa Cruz.....	18.3	0 <sup>b</sup>	27.6	17.2 <sup>b</sup>	20.9 <sup>b</sup>	33.7
Santa Rosa.....	31.9	31.6	0 <sup>b</sup>	72.2	16.6 <sup>b</sup>	55.2
Scotia.....	0 <sup>b</sup>	0 <sup>b</sup>	0 <sup>b</sup>	50.5	14.5 <sup>b</sup>	11.1 <sup>b</sup>
Tahoe City.....			58.2	55.4	31.5	
Vallejo.....						
Fleming Hill.....	14.0	55.4	46.1	60.6	48.6	
Swanzy Reservoir.....	0 <sup>b</sup>	29.3	34.3	59.1	23.0	
Vista.....	0 <sup>b</sup>	31.2	26.8			27.2
Willits.....				103.7	33.8	29.5
Yosemite.....	0 <sup>b</sup>	110	24.0	72.4	54.5	143.0

<sup>a</sup> Blank space indicates no sample collected or analyzed.

<sup>b</sup> The counting rate of the sample is not equal to at least twice the 0.95 statistical counting error but the value reported is the best available estimate.

<sup>c</sup> Average of more than one sample for the month.

ary through June 1963. Following treatment, these waters are used for industrial and domestic purposes. Because alpha activity in water has, in general, been undetectable or very slight, alpha activity analyses are not presented. Little increase in the radioactivity level of surface water is observed in spite of some increase in fallout.

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Previous coverage in *Radiological Health Data*:

Period	Issue
1961—June 1962	April 1963
July—December 1962	September 1963

## Section IV.—Other Data

### IODINE-131 IN POST-MORTEM HUMAN THYROIDS

Frances I. Visalli and Abraham S. Goldin<sup>1</sup>

The iodine-131 burdens of post-mortem thyroids from New England residents coming to autopsy at two lying-in hospitals and one children's hospital were measured at the Northeastern Radiological Health Laboratory from May 1962 to January 1963. Nuclear weapons tests were conducted preceding and during this same period by the United States and by the U. S. S. R. and the following results are observations made on available thyroid tissue during that period. Practically all of the samples were obtained from peri- or pre-natal deaths, or as a result of deaths of infants or children from disease. The population represented can therefore not be considered as typical.

#### Method of Measurement

The method of measurement used was gamma scintillation spectroscopy. The thyroid sample as received in a small plastic vial was placed on a 4 x 4-inch sodium iodide crystal (thallium activated) inside a 6-inch-thick iron shield. The detector was connected to a multi-channel analyzer and the iodine-131 content

calculated from the counting rate in the region from 0.33 to 0.39 Mev after correction for background. This is essentially the method used by Eisenbud *et al.* (1). There was no measurable contribution in these small samples from potassium-40 or cesium-137. With this equipment, in a 50-minute count, the measured  $2\sigma$  counting error for samples of low radioiodine content was approximately 2.6 counts per minute, corresponding to 9 picocuries. After these values were corrected for radioactive decay from the time of death to the time of measurement, the  $2\sigma$  error was usually about  $\pm 13$  to 17 picocuries for the entire sample. In a few cases, where a relatively long period intervened between the time of death and the time of measurement, the error was greater than the above range. The minimum detectable activity is defined as the smallest value at which the confidence interval, the lower limit of which is established by the observed value minus 2 standard deviations due to counting, remains above zero. Since many of the samples were quite small, the error, when expressed as picocuries per gram, becomes numerically large when compared to the sample value.

#### Results

Since both the gross sample rate and the background rate are subject to random statis-

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tical variations, the difference between them (net rate) may be less than zero when samples of zero or very low activity are being measured. The actual values, whether positive or negative, are given in table 1. The activity of individual samples should be considered "not detectable" when the reported value is negative or is less than the reported counting error, but the actual values should be used in averaging.

The results of iodine-131 measurements made on thyroids from persons in New England between May 1962 and January 1963 are shown in table 1.

In nearly all of these cases, the thyroid was removed for *post-mortem* examination and the portion submitted was that part of the thyroid not required by the pathologist in his examination. Information as to total thyroid size or

TABLE 1.—IODINE-131 IN POST MORTEM THYROID

Sample number	Date of death	Age at death	Sex	Weight (pounds)	Weight of sample submitted (grams) <sup>a</sup>	Iodine-131 in sample <sup>b</sup>	
						(pc)	(pc/g)
GROUP I							
0-100	5/14/62	13 yr 11 mo	F		2.7	0 ± 14	0 ± 5
0-101	5/10/62	2 yr 5 mo	M		0.5	<sup>c</sup> -2 ± 21	<sup>c</sup> -4 ± 42
0-102	5/15/62	4 yr 2 mo	F	44	1.0	9 ± 15	9 ± 15
0-103	5/16/62	2.5 mo	M	15	0.4	8 ± 14	20 ± 35
0-104	5/15/62	7 yr 3 mo	F	43	8.5	430 ± 29	51 ± 3.4
0-105	5/23/62	24 hrs.	M	2	0.5	<sup>c</sup> -12 ± 14	<sup>c</sup> -27 ± 31
0-106	5/25/62	3 yr 9 mo	F	40	2.6	<sup>c</sup> -3 ± 16	<sup>c</sup> -1.2 ± 6.1
0-107	5/27/62	10 days	F	6.2	0.6	0.4 ± 14	0.7 ± 23
0-108	5/30/62	13 yr 7 mo	M	117	6.4	7 ± 16	1.1 ± 2.5
0-109	5/30/62	1 yr 11 mo	M	25	0.5	<sup>c</sup> -5 ± 16	<sup>c</sup> -10 ± 32
0-110	5/30/62	8 yr 8 mo	M	48	5.4	15 ± 16	2.8 ± 3.0
0-111	5/30/62	14 years	F	132	3.9	2 ± 16	0.5 ± 4.1
0-112	6/ 3/62	3 yr 11 mo	F	44	1.6	5 ± 13	3.1 ± 8.1
0-113	6/ 5/62	4 yr 7 mo	F	30	1.9	7 ± 16	3.7 ± 8.4
0-114	6/ 8/62	1 mo	M	2	0.3	4 ± 12	13 ± 40
0-115	6/11/62	4 mo	F	13	0.4	6 ± 12	15 ± 30
0-117	6/16/62	7 yr 1 mo	M	60	1.7	<sup>c</sup> -27 ± 27	<sup>c</sup> -16 ± 16
0-118	6/23/62		M	4.6	0.7	<sup>c</sup> -12 ± 16	<sup>c</sup> -17 ± 23
0-119	6/22/62	20 years	F	100	3.1	<sup>c</sup> -4 ± 17	<sup>c</sup> -1.3 ± 5.5
0-120	6/23/62	23 days	F	3	0.6	3 ± 16	5 ± 27
0-121	6/25/62	5 days	M	7.6	0.4	<sup>c</sup> -5 ± 13	<sup>c</sup> -12 ± 33
0-122	6/27/62	6 yr 10 mo	F	43	1.3	<sup>c</sup> -7 ± 12	<sup>c</sup> -5.4 ± 9.2
0-123	6/29/62	4 yr 4 mo	M	35	1.1	<sup>c</sup> -7 ± 10	<sup>c</sup> -6.4 ± 9.1
0-124	6/28/62	31 hours	M	4.8	0.5	<sup>c</sup> -4 ± 11	<sup>c</sup> -8 ± 22
0-125	7/ 2/62	30 hours		6.1	0.6	12 ± 20	20 ± 33
0-126	7/ 2/62	38 hours	F	3.4	0.2	3 ± 20	15 ± 100
0-130	7/10/62	12 hours	F	2.6	0.4	7 ± 15	18 ± 38
0-131	7/10/62	5 hours	F	2.4	0.4	<sup>c</sup> -2 ± 15	<sup>c</sup> -5 ± 38
0-132	7/11/62	1 yr 6 mo	F	22	0.9	12 ± 18	13 ± 20
0-133	7/17/62	1 mo	F	7	0.5	4 ± 11	8 ± 22
0-134	7/17/62	13 min	M	4.6	0.4	<sup>c</sup> -2 ± 20	<sup>c</sup> -5 ± 50
0-135	7/20/62	1 mo	F	4.2	0.1	2 ± 15	20 ± 150
0-136	7/24/62	4 ½ hrs	M	5.3	0.7	20 ± 11	29 ± 16
0-137	7/24/62	Stillborn	M	7.4	0.3	8 ± 11	27 ± 37
0-138	7/25/62	15 yr 7 mo	M	103	10.9	51 ± 19	4.7 ± 1.7
0-139	8/21/62	8 yr 6 mo	M	85	2.7	1 ± 15	0.4 ± 5.6
0-140	8/30/62	4 mo	F	11.5	0.5	<sup>c</sup> -2 ± 26	<sup>c</sup> -4 ± 52
Average of 37 samples						4.3	
GROUP II							
0-141	9/20/62	24 hrs	F	1.5	0.3	0 ± 17	0 ± 57
0-142	9/15/62	5 yr 9 mo	M	48	2.3	4 ± 25	2 ± 11
0-143	9/21/62	6 years	M	50	3.9	16 ± 15	4.1 ± 3.9
0-144	9/25/62	34 years	M		7.0	5 ± 17	0.7 ± 2.4
0-145	10/10/62	18 days	M	6.3	0.6	7 ± 30	12 ± 50
0-146	10/15/62	Stillborn	M	7.2	0.8	1 ± 22	1 ± 28
0-147	10/15/62	Stillborn	F	1.5	0.4	6 ± 19	15 ± 48
0-148	10/17/62	Stillborn	F	2.5	0.4	0 ± 18	0 ± 45
0-149	10/17/62	2 mo	M	10	0.7	9 ± 17	13 ± 24
0-150	10/22/62	15 yr 7 mo	M	124	7.4	118 ± 15	16 ± 2
0-151	10/24/62	11 hrs	F	4.9	0.5	23 ± 11	46 ± 22
0-152	10/24/62	11 hrs	M	4.4	0.8	14 ± 10	18 ± 13
0-153	10/26/62	45 hrs	M	5.2	0.6	35 ± 13	58 ± 22
0-154	10/26/62	13¾ hrs	F	6.3	1.0	30 ± 12	30 ± 12
0-155	10/21/62	Stillborn	F	7.6	0.7	15 ± 41	22 ± 59
0-156	11/ 4/62	2 hrs	F	4.2	0.9	8 ± 13	9 ± 14
0-157	12/13/62	22¾ hrs	F	9.8	0.8	7 ± 29	9 ± 36
0-158	12/14/62	5 days	F	4	0.2	0 ± 52	0 ± 260
0-159	12/21/62	3 mo	F	10.8	1.3	0 ± 25	0 ± 19
0-160	1/10/63	Stillborn	F	5.6	0.8	23 ± 18	29 ± 22
Average of 20 samples						14.2	

<sup>a</sup> The weight of sample submitted does not constitute the entire thyroid mass in all cases.

<sup>b</sup> Corrected to date of death; error shown is the 2σ statistical counting error.

<sup>c</sup> Negative values arise from subtracting statistically fluctuating background. (see Results above).

total thyroid burden is not available. All the values provided should be considered as representing only that portion of the thyroid furnished.

Environmental levels of radioiodine in New England, as measured in market milk (2) and in institutional diet samples (3) were near or below the limits of detectability from May 1962 until about September 15, 1962. In addition, air particulate levels did not indicate the presence of fresh fission products (4). Thus, the data of table 1 may be best considered in two groups. Group I consists of 37 samples, numbered 0-100 through 0-140, which were collected when the environmental levels were relatively low. There are three samples in Group I in which the measured activity was greater than the  $2\sigma$  error, indicating the presence of radioactive iodine. In one case (0-104) the level is quite high and examination of the spectrum obtained indicated that iodine-131 was definitely present. The source of this radioiodine is unknown; neither the administration of radioiodine nor its non-administration could be definitely established. There was also an indication of iodine-131 in samples 0-136 and 0-138, although at a substantially lower level. In the other 34 samples in this group, the levels were such that in no case was there a definite indication of the presence of detectable quantities of radioiodine.

Samples numbered 0-141 through 0-160 form Group II. These were collected when the environmental iodine-131 levels were elevated. Of these 20 samples, seven contained iodine-131 in quantities greater than the error of observation. The average for this group of 20 samples is 14.2 picocuries per gram; the average for the seven which are significantly greater than zero is 28.7 picocuries per gram. In contrast, the average for the first 37 samples (excluding number 0-104) is 3.0 picocuries per gram.

### Discussion

Except as mentioned above, iodine-131 burdens of thyroid samples obtained from individuals who died before the middle of September 1962 were not significantly different from zero, reflecting the absence of measurable iodine-131 in the environment. The positive values

noted from the middle of September 1962 to the end of October 1962 reflect the readily measurable environmental iodine-131 levels during that period. After the end of October 1962, iodine-131 concentrations in New England milk samples (2) began to recede. However, a drop in iodine-131 for the institutional diet sample (3) did not occur until after December. Since only five samples were obtained during this period, no conclusions can be drawn from them.

It may also be of interest to compare the observed iodine-131 levels in thyroids with appropriate radiation protection standards. Using the data of the International Commission on Radiological Protection (5), the dose to the thyroid from iodine-131 is 0.082 mrem/wk per pc/g of thyroid. On a yearly basis this is 4.3 mrem/yr per pc/g. A thyroid concentration of about 350 picocuries of iodine-131 per gram, maintained over a year, would therefore cause a dose equal to the Federal Radiation Council's Radiation Protection Guide (RPG) (6) of 1.5 rem/yr for individuals in an exposed population. Similarly, an average thyroid concentration of about 115 picocuries of iodine-131 per gram, maintained over a year, would produce the RPG (6) of 0.5 rem/yr to the average of an exposed population. The highest individual value in table 1, 58 pc/g, if sustained for a period of time on the order of one year, would represent about one-sixth of the RPG of 1.5 rem/yr for *individuals*. Similarly, the average of the 20 samples in Group II, 14.2 pc/g, if sustained for one year, would represent about one-eighth of the RPG of 0.5 rem per year for an average of an *exposed population*.

While these measurements of the iodine-131 content of *post-mortem* thyroids represent direct measurements of the presence of this nuclide in an extremely limited portion of a population, caution should be taken in extending the data beyond the actual samples involved. Since these thyroids were obtained from a moribund population, the dietary intake and metabolism of iodine-131 may be far different from those existing in the general population. Thus, the thyroid burdens of iodine-131 in hospitalized patients cannot be assumed to be representative of those existing in the population at large.

### Summary.

Fifty-seven thyroid samples were made available by three New England hospitals during the period May 14, 1962 through January 10, 1963. Up to the middle of September, iodine-131 was detected in three out of 37 samples (range 47 to 51 pc/gram). Positive values for iodine-131 were found in 6 out of 15 *post-mortem* thyroids from September 15 through October 26 (range 41 to 58 pc/gram). From November 4 through January 10, 1963, one positive value (29 pc/gram) of iodine-131 was noted among 5 samples collected. The highest individual thyroid burden noted represented about one-sixth of the RPG of 1.5 rem/yr, and the average of 20 samples after September 15, 1962 represented about one-eighth of the RPG of 0.5 rem/yr.

### Acknowledgements

The measurements of iodine-131 in the thyroids were performed by the Northeastern Radiological Health Laboratory's Analytical Services Section, under the direction of Mr.

Edmond J. Baratta. Mr. Robert P. Chandler, Technical Editor, Radiological Health Data & Reports Staff, assisted in the preparation of the manuscript.

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## ENVIRONMENTAL LEVELS OF RADIOACTIVITY AT ATOMIC ENERGY COMMISSION INSTALLATIONS

The U. S. Atomic Energy Commission receives from its contractors quarterly reports on the environmental levels of radioactivity in the vicinity of major Commission installations. The reports include data from routine monitoring programs where operations are of such a nature that plant perimeter surveys are required.

Summaries of the environmental radioactivity data for 22 AEC installations have appeared periodically in *Radiological Health Data* since November 1960. A summary follows for Shippingport Atomic Power Station.

Releases of radioactive materials from the Shippingport Plant are regulated in accordance with standards set forth in the Federal Register Title 10—Part 20. The appropriate concentration standards are given in table 1.

### Shippingport Atomic Power Station, July 1962-June 1963

*Duquesne Light Company, Shippingport, Pennsylvania*

Environmental radiation monitoring at the Shippingport Atomic Power Station began with a two-year preoperational survey program to establish background levels at the site of the world's first large-scale nuclear-powered electric generating station. Following initial operation of the plant in December 1957, this program was continued as originally conceived through the third quarter of 1961, when it was determined that fewer sampling locations closer to the plant would provide equal or better evaluation of the effects of plant operation on the environment.

#### *Release of Radioactive Materials to the Atmosphere*

Controlled releases of xenon-133 were made periodically at a concentration of 130,000 pc/m<sup>3</sup> at the stack. A total of 12.1 mc Xe<sup>133</sup> were discharged during the second half of 1962 and 406 mc during the first half of 1963.

TABLE 1.—MAXIMUM PERMISSIBLE CONCENTRATIONS OF EFFLUENTS DISCHARGED TO THE SHIPPINGPORT PLANT ENVIRONMENT\*

Radionuclide	Water (pc/liter)	Air (pc/m <sup>3</sup> )
Hydrogen-3 (tritium).....	3,000,000	—
Xenon-133.....	—	300,000
Mixture of unidentified radionuclides.....	10	—
Mixture, if Ra <sup>226</sup> and Ra <sup>228</sup> are not present <sup>b</sup> .....	100	—
Mixture, if Sr <sup>90</sup> , I <sup>131</sup> , Pb <sup>210</sup> , Ac <sup>227</sup> , Ra <sup>226</sup> , Pa <sup>230</sup> , Pu <sup>241</sup> , Bk <sup>249</sup> are not present <sup>b</sup> .....	—	100

\* The concentration standards given here were taken from The Atomic Energy Commission's regulation 10 CFR Part 20 (Federal Register, November 17, 1960).

<sup>b</sup> "Not present" implies the concentration of the nuclide is small compared with its appropriate MPC. According to AEC regulation (Federal Register, Title 10, Part 20, August 9, 1961), a group of nuclides may be considered not present if the ratio of each nuclide to its appropriate MPC is equal to or less than 1/10 and if the sum of these ratios for the group in question is equal to or less than 1/4.

The exhaust from an incinerator for burning contaminated combustible material is passed through a wet scrubber and a high efficiency filter before entering the stack. The average gross radioactivity of incinerator effluent during operation was 27 pc/m<sup>3</sup> during the second half 1962 and 18 pc/m<sup>3</sup> during the first half 1963.

#### *Air Monitoring*

Airborne particulates are sampled at three area monitoring stations by means of a continuously moving paper tape sampler with an end-window Geiger-Mueller detector and recorder. A one-half hour decay time is allowed before counting, hence, naturally-occurring radon and thoron daughters would be present. Six month average activities are given in table 2. Area monitoring stations 1, 2, and 3 are located 150 yards southeast, 150 yards west and one-half mile north-northwest of the stack, respectively.

TABLE 2.—GROSS BETA IN AIR

[Average concentrations in pc/m<sup>3</sup>]

Station number	Second half 1962	First half 1963
1-----	1.0	1.0
2-----	not operating	1.3
3-----	1.5	1.4

*Fallout Sampling*

Monthly pot samples were collected at each of the three area monitoring stations (same locations as for air). Gross beta average monthly deposition rates are given in table 3.

*Liquid Radioactive Waste Disposal*

Continuous controlled discharges of tritium and other unidentified radioactive wastes are made into the Ohio River. The quantities of discharge and average effluent concentration are given in table 4.

*Ohio River Water Monitoring*

River water samples were collected by a continuous samples upstream at the Shippingport condenser cooling water intake and by grab sampling downstream at the condenser cooling water outfall. (The downstream continuous sampler was not functioning.) These samples

TABLE 3.—GROSS BETA IN FALLOUT

[Average monthly deposition in nc/m<sup>2</sup>]

Station number	Second half 1962	First half 1963
1-----	199	276
2-----	209	340
3-----	193	372

**REPORTED NUCLEAR DETONATIONS, FEBRUARY 1964**

One nuclear detonation was announced by the U. S. Atomic Energy Commission during February 1964. This test was of low intermediate (20 to 200 kilotons) yield and was conducted underground at the Nevada Test Site on February 20 as part of the Commission's

TABLE 4.—LIQUID WASTES DISCHARGED INTO THE OHIO RIVER

	Tritium		Unidentified radioactivity	
	Second half 1962	First half 1963	Second half 1962	First half 1963
Total discharge (mc)	621	1,381	51	106
Average of monthly average concentrations of effluent (pc/liter)	172	61	1.8	4.4

were analyzed weekly for both alpha and beta, suspended and dissolved radioactivity. The six month averages of these measurements are given in table 5.

TABLE 5.—RADIOACTIVITY IN OHIO RIVER WATER

[Average concentrations in pc/liter]

	Location	Alpha		Beta	
		Second half 1962	First half 1963	Second half 1962	First half 1963
Suspended	Upstream	0.13	8.79	5.0	33.8
solids	Downstream	0.14	1.24	7.8	31.8
Dissolved	Upstream	0.86	0.86	24.6	25.3
solids	Downstream	0.64	1.23	28.8	32.3

*Previous coverage in Radiological Health Data:*

Period	Issue
1959	July 1960
First quarter 1960	December 1960
Second quarter 1960	January 1961
Third and fourth quarters 1960	October 1961
First and second quarters 1961	April 1962
Second half 1961 and first half 1962	May 1963

"Plowshare Program" to develop peaceful uses for nuclear explosives. The test was one of a series to develop devices for use in possible later excavation experiments. Arbitrary reference number 150 was assigned by *Radiological Health Data*.

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**UNITED STATES GOVERNMENT PRINTING OFFICE, WASHINGTON 25, D.C. 1961**

**For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C.**

**Subscription price \$5.00 a year, \$1.50 additional for foreign mailing.**

**Price for a single copy of this issue is 50 cents.**

March 1964



# UNITS AND EQUIVALENTS

Symbol	Unit	Equivalent
Bev.....	billion electron volt	
cpm.....	count per minute	
dpm.....	disintegration per minute	
g.....	gram	
kg.....	kilogram	1 kg = 1000 gm = 2.2 pounds
km <sup>2</sup> .....	square kilometer	
kvp.....	kilovolt peak	
m <sup>3</sup> .....	cubic meter	1 m <sup>3</sup> = 1000 liters
ma.....	milliampere	
mas.....	milliampere-second	
Mev.....	million electron volts	
mi <sup>2</sup> .....	square mile	
ml.....	milliliter	
mm.....	millimeter	
mrad.....	millirad	
mrem.....	millirem	
mr/hr.....	milliroentgen per hour	
mμc.....	millimicrocurie	1 mμc = 1 nc
nc.....	nanocurie	1 nc = 1000 pc = 1 mμc = 10 <sup>-9</sup> curies
nc/m <sup>2</sup> .....	nanocurie per square meter	1 nc/m <sup>2</sup> = 1 mμc/m <sup>2</sup> = 1,000 μμc/m <sup>2</sup> = 1 mc/km <sup>2</sup> = 2.59 mc/mi <sup>2</sup>
pc.....	pico curie	1 pc = 1 μμc = 10 <sup>-12</sup> curies
r.....	roentgen	
μμc.....	micromicrocurie	1 μμc = 2.22 dpm

# INTERNATIONAL NUMERICAL MULTIPLE AND SUBMULTIPLE PREFIXES

Multiples and submultiples	Prefixes	Symbols	Pronunciations
10 <sup>12</sup>	tera	T	tēr' a
10 <sup>9</sup>	giga	G	jī' ga
10 <sup>6</sup>	mega	M	mēg' a
10 <sup>3</sup>	kilo	k	kīl' o
10 <sup>2</sup>	hecto	h	hēk' to
10	deka	da	dēk' a
10 <sup>-1</sup>	deci	d	dēs' i
10 <sup>-2</sup>	centi	c	sēn' tī
10 <sup>-3</sup>	milli	m	mīl' i
10 <sup>-6</sup>	micro	μ	mī' kro
10 <sup>-9</sup>	nano	n	nān' o
10 <sup>-12</sup>	pico	p	pē' co
10 <sup>-15</sup>	femto	f	fēm' to

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